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THÈSE DE DOCTORAT

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Toan Khanh VU

Polluants organiques persistants ubiquistes et historiques à contemporains dans le fleuve Rouge, de Hanoï au golfe du Tonkin (Vietnam) : présence, distribution spatiale et saisonnière, et évaluation des risques

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Affidavit

I, undersigned, Toan Khanh VU, hereby declare that the work presented in this manuscript is my own work, carried out under the scientific supervision of *Laure MALLERET, Vincent FAUVELLE, and Van Hoi BUI*, in accordance with the principles of honesty, integrity and responsibility inherent to the research mission. The research work and the writing of this manuscript have been carried out in compliance with both the French national charter for Research Integrity and the Aix-Marseille University charter on the fight against plagiarism.

This work has not been submitted previously either in this country or in another country in the same or in a similar version to any other examination body.

À Toulouse, 10 octobre 2025.



Toan Khanh VU.

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Liste de publications et/ou brevets et participation aux conférences

1) Liste des publications et/ou brevet réalisées dans le cadre du projet de thèse :

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Abstract

Vietnam, a rapidly developing country in Southeast Asia, has experienced strong demographic and economic growth in recent decades, with the Gross Domestic Product (GDP) growth exceeding 7% in 2024 and a population surpassing 100 million. Such growth has driven urbanization, industrialization, and increasing imports, thus intensifying environmental pressures, and particularly the chemicals' inputs. Despite regulatory progress, including national technical regulations and Vietnam's commitment to the Stockholm Convention on Persistent Organic Pollutants (POPs), wastewater treatment and chemicals' management remain limited. Thus, many hazardous organic contaminants may be released into aquatic systems through industrial and urban discharges, agricultural runoff, and insufficient treatment abatement.

This PhD project aims to explore how this rapid socio-economic development is impacting the quality of aquatic environments, exploring the Red River (the largest river in northern Vietnam, flowing through Hanoi capital), as a typical case study. The research focuses on three groups of organic contaminants sweeping across different regulatory and knowledge contexts: ubiquitous and well-studied ubiquitous priority contaminants (polycyclic aromatic hydrocarbons, PAHs), widely used as benchmarks for assessing environmental quality; historical POPs (polychlorinated biphenyls, PCBs, polybrominated diphenyl ethers, PBDEs, and chlorinated paraffins, CPs), all listed for their persistence, long-range transport, toxicity and bioaccumulation, and for which regulation has progressively evolved, with some compounds only recently listed (e.g., medium-chain chlorinated paraffins in 2025); and per- and polyfluoroalkyl substances (PFAS), a large and diverse group including both already-regulated POPs and emerging, understudied compounds.

The doctoral research work addresses the general question of how such contaminants can reveal the impact of demographic and economic growth on aquatic environments. Specifically, it i) examines how they differ in terms of regulatory status, environmental persistence, and data availability, ii) investigates their concentrations and spatial-seasonal distribution patterns in Red River water and sediment samples, and iii) assessed the risk caused to aquatic organisms.

Generating such baseline data is essential to fill existing knowledge gaps in Vietnam's monitoring and regulatory requirements. The findings aim to support the development of national strategies for sediment monitoring and bring data to help in building environmental risk assessment frameworks in Vietnam.

Keywords: Vietnam, Red River, Gulf of Tonkin, PAHs, PCBs, PBDEs, CPs, PFAS, sediment, water.

Résumé

Le Vietnam, pays en plein essor situé en Asie du Sud-Est, a connu une forte croissance démographique et économique au cours des dernières décennies, avec une croissance du produit intérieur brut (PIB) supérieure à 7 % en 2024 et une population dépassant les 100 millions d'habitants. Cette croissance a entraîné l'urbanisation, l'industrialisation et l'augmentation des importations, intensifiant ainsi les pressions environnementales, en particulier celles liées à l'utilisation de produits chimiques. Malgré les progrès réglementaires, notamment les réglementations nationales et la ratification par le Vietnam de la Convention de Stockholm sur les polluants organiques persistants (POP), le traitement des eaux usées et la gestion des produits chimiques restent limités. Ainsi, de nombreux contaminants organiques dangereux peuvent être liberés dans les systèmes aquatiques par les rejets industriels et urbains, le ruissellement agricole et le manque d'infrastructures de traitement.

Ce projet de doctorat vise à étudier l'impact de ce développement socio-économique rapide sur la qualité des milieux aquatiques, en prenant comme étude de cas typique le fleuve Rouge (le plus grand fleuve du nord du Vietnam, qui traverse la capitale Hanoï). La recherche se concentre sur trois groupes de contaminants organiques qui recouvrent différents contextes réglementaires et scientifiques : des contaminants prioritaires ubiquistes et bien étudiés (hydrocarbures aromatiques polycycliques, HAP), largement utilisés comme comparateurs pour évaluer la qualité des milieux ; les POP règlementés (polychlorobiphényles, PCB, polybromodiphényléthers, PBDE et paraffines chlorées, PC), tous répertoriés pour leur persistance, leur transport à longue distance, leur toxicité et leur bioaccumulation, et pour lesquels la réglementation a progressivement évolué, certains composés n'ayant été répertoriés que récemment (par exemple, les paraffines chlorées à chaîne moyenne en 2025) ; et les substances per- et polyfluoroalkylées (PFAS), un groupe vaste et diversifié comprenant à la fois des POP déjà réglementés et des composés émergents peu étudiés.

Les travaux de recherche doctorale questionnent l'utilisation de ces contaminants comme marqueurs de l'empreinte anthropique susceptible de révéler l'impact de la croissance démographique et économique sur les milieux aquatiques. Plus précisément,

les recherches i) examinent les statuts réglementaires, la persistance dans l'environnement et la disponibilité des données d'occurrence de ces différents contaminants, ii) évaluent leurs concentrations et leurs distributions spatiales et saisonnières dans des échantillons d'eau et de sédiments prélevés dans le fleuve Rouge, et iii) estiment les risques qu'ils présentent pour les organismes aquatiques.

La production de ces données est nécessaire pour combler les lacunes existantes en matière de surveillance et de réglementation au Vietnam. Les résultats visent à soutenir l'élaboration de stratégies nationales de surveillance des sédiments et à fournir des données utiles à la mise en place de cadres d'évaluation des risques environnementaux au Vietnam.

Mots clés: Vietnam, Fleuve Rouge, Golfe de Tonkin, HAPs, PCBs, PBDEs, CPs, PFAS, sédiment, eau.

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Résumé étendu

Le Vietnam, pays en plein essor situé en Asie du Sud-Est, a connu une forte croissance démographique et économique au cours des dernières décennies, avec une croissance du produit intérieur brut (PIB) supérieure à 7 % en 2024 et une population dépassant les 100 millions d'habitants. Cette croissance a entraîné l'urbanisation, l'industrialisation et l'augmentation des importations, intensifiant ainsi les pressions environnementales, en particulier celles liées à l'utilisation de produits chimiques. Malgré les progrès réglementaires, notamment les réglementations techniques nationales et l'engagement du Vietnam envers la Convention de Stockholm sur les polluants organiques persistants (POP), le traitement des eaux usées et la gestion des produits chimiques restent limités. Ainsi, de nombreux contaminants organiques dangereux pour l'homme ou son environnement peuvent être déversés vers les systèmes aquatiques par les rejets industriels et urbains, le ruissellement agricole et l'insuffisance des traitements de dépollution. Ceci amène à se questionner sur l'impact de ces développements sur l'état des milieux aquatiques au Vietnam et dans quelle mesure cet impact peut être mis en évidence pour le diagnostic des pollutions.

Cette thèse de doctorat vise à étudier l'impact du développement socio-économique rapide du Vietnam, sur la qualité des milieux aquatiques, en prenant comme étude de cas typique le fleuve Rouge. Le fleuve Rouge prend sa source dans le sud de la Chine et traverse le nord du Vietnam, passant par Hanoï, la capitale densément peuplée du Vietnam, ainsi que les zones industrielles et agricoles du delta du fleuve Rouge, avant de se jeter dans le golfe du Tonkin. La combinaison des rejets industriels, des eaux usées domestiques, des ruissellements agricoles et de l'urbanisation rapide a fait de ce fleuve un point chaud pour les pressions anthropiques.

Les travaux se sont intéressés à cinq familles de contaminants organiques qui transcendent différents contextes réglementaires et scientifiques. Les hydrocarbures aromatiques polycycliques (HAP) ont été sélectionnés car il s'agit de contaminants prioritaires ubiquistes et déjà bien étudiés dans différents milieux servant d' échelle d'anthropisation, , car ils sont déjà largement utilisés comme références pour évaluer la qualité des environnements. Les quatre autres familles de contaminants sont des

polluants organiques persistants (POP), ou polluants dits éternels, car ils persistent de longues années dans l'environnement, suite à leurs rejets. Ils présentent des caractéristiques de persistance, transport à longue distance, toxicité et sont susceptibles de se bioaccumuler. Les polychlorobiphényles (PCB) sont d'usage historiques et ont été réglementés par la convention de Stockholm dès sa mise en place. Les polybromodiphényléthers (PBDE) et paraffines chlorées à chaines courtes et moyennes(CPs) ont été commercialisés par la suite, parfois en remplacement des PCBs pour certaines applications et ont été inclus progressivement dans la convention de Stockholm entre 2009 et 2025 (dans le cas des paraffines chlorées à chaîne moyenne) tous répertoriésEnfin les substances per- et polyfluoroalkylées (PFAS), un groupe vaste et diversifié de molécules, comprennent à la fois des POP déjà réglementés et des composés émergents encore peu étudiés. Par cette approche multi-molécules faiblement dégradables, nous voulons explorer leur pertinence pour évaluer à quel point le milieu est atteint et dans quelle mesure ces atteintes reflètent les restrictions réglementaires et les successions entre mises sur le marché et retrait.

Dans le contexte législatif et économique présenté ci-dessus, l'objectif général de cette thèse est de renforcer les connaissances actuelles sur la présence et les comportements des HAP, des PCB, des PBDE, des CP et des PFAS dans l'environnement aquatique vietnamien, et plus précisément dans le fleuve Rouge situé au nord du Vietnam.

Outre un objectif spécifique de la recherche, axé sur l'examen de l'état actuel des connaissances concernant les composés étudiés dans l'environnement aquatique d'Asie du Sud-Est, l'objectif opérationnel spécifique de mon travail de recherche était de combler les lacunes sur la situation de contamination du fleuve Rouge en étudiant la présence et la dynamique de cinq groupes de contaminants organiques dans ce système hydrographique. Cet objectif opérationnel spécifique a été décomposé et traité en trois sous-objectifs : 1) Etendre la couverture spatiale et temporelle des mesures des HAP, fournir une identification affinée des sources et évaluer les variations saisonnières ainsi que les risques potentiels associés ;

2) S'intéresser à l'occurrence des PCB, PBDE et CP dans les sédiments de la rivière Rouge en générant un ensemble de données originales, en identifiant les sources de contamination possibles et en examinant le comportement de ces composés le long du fleuve et en fonction des saisons.

3) Adapter et valider la méthode d'extraction basée sur la norme US-EPA1633 et appliquer à la fois une analyse ciblée et une quantification globale (c'est-à-dire le dosage des précurseurs oxydables totaux, TOPA) dans l'eau et les sédiments de la rivière Rouge, afin d'étudier un large éventail de PFAS, y compris la partie « cachée » des PFAS qui n'aurait pas été couverte par l'approche ciblée

Pour atteindre ces objectifs opérationnels, quatre campagnes d'échantillonnage ont été menées dans le fleuve Rouge (transect de 200 km entre le centre-ville de Hanoï et l'estuaire de Ba Lat) de novembre 2022 à septembre 2023. Plusieurs méthodes d'extraction (extraction liquide sous pression et extraction assistée par ultrasons) et d'analyse (chromatographie en phase gazeuse et liquide couplée à la spectrométrie de masse) ont été utilisées, afin de quantifier les cinq groupes de contaminants organiques.

Après une introduction générale, la thèse s'articule en cinq chapitres, qui sont rédigés au format publication. Le chapitre 1 comprend une introduction du contexte réglementaire international et des informations générales sur les familles chimiques d'intérêt. Il présente également une revue systématique de la présence de HAP au Vietnam et des POP industriels dans le milieu aquatique, non seulement au Vietnam, mais dans toute l'Asie du Sud-Est.

Au cours des deux dernières décennies, de nombreuses études ont mis en évidence une distribution généralisée des hydrocarbures aromatiques polycycliques (HAP) dans les sédiments vietnamiens. Les concentrations les plus élevées sont systématiquement observées dans les cours d'eau urbains et industriels, notamment dans les deux principales métropoles que sont Hanoï et Hô Chi Minh-Ville, tandis que les zones rurales et côtières présentent généralement des teneurs plus faibles. La prédominance des HAP de masse moléculaire moyenne à élevée, rapportée dans la majorité des travaux, traduit une contribution majeure de sources pyrogéniques ainsi qu'une forte persistance dans les matrices sédimentaires. Si des critères de qualité des sédiments ont été définis, l'absence de normes réglementaires encadrant les HAP dans les eaux de surface vietnamiennes met en lumière la nécessité de développer des programmes de surveillance intégrés et de renforcer les mesures de gestion environnementale.

Les PCB, PBDE, CP et PFAS constituent quatre groupes de produits chimiques industriels inscrits par la Convention de Stockholm, non pas en tant que composés individuels mais en tant que groupes de contaminants comprenant de nombreuses molécules. L'examen de ces quatre POPs dans les milieux aquatiques d'Asie du Sud-Est

(Publication N°1) met en évidence un défi majeur et persistant en matière de gestion environnementale. Malgré les efforts déployés pour atteindre les objectifs de la Convention de Stockholm, les concentrations de ces POPs dans les ressources en eau de la région n'ont pas montré de tendance claire à la baisse au cours des trente dernières années. Cette persistance est largement attribuée à des rejets continus provenant de sources actuelles. La présence de ces POPs dans les cours d'eau urbains, les environnements marins, les organismes aquatiques, ainsi que dans l'eau du robinet et de consommation, souligne le risque d'exposition humaine par l'ingestion d'eau et d'aliments contaminés.

Au bilan, la littérature scientifique sur l'Asie du Sud-Est, bien qu'en expansion, présente encore d'importantes lacunes géographiques et spécifiques aux composés. Aucune donnée sur les CPs n'est disponible dans les milieux aquatiques des onze pays documentés, et aucun suivi des quatre groupes de composés n'a été recensé au Brunei, au Myanmar et au Timor-Leste, probablement en raison de contextes socio-économiques complexes.

Il est donc crucial de renforcer et de coordonner les efforts de suivi environnemental dans la région. Les recherches futures doivent combler ces lacunes pour mieux caractériser la contamination régionale, en particulier face à l'urbanisation rapide et aux phénomènes climatiques extrêmes, afin d'informer efficacement les politiques et stratégies de gestion des ressources en eau.

Le chapitre 2 présente succinctement une vue d'ensemble de la zone d'étude, des campagnes d'échantillonnage, ainsi que des méthodes d'extraction et d'analyse utilisées dans cette thèse et qui sont ensuite explicités de façon plus précise dans chaque chapitre de résultats.

Le chapitre 3 traite des variations spatio-temporelles des HAP détectés dans les échantillons de sédiments du fleuve Rouge, ainsi que de l'évaluation de leurs sources et des risques associés (Publication N°2).

Les concentrations totales des 16 HAP prioritaires dans les sédiments du fleuve Rouge variaient de 16,2 à 57,9 ng g^{-1} pendant la saison sèche (mars 2023) et augmentaient durant la saison des pluies, atteignant 94,7 ng g^{-1} en juin et 108,0 ng g^{-1} (septembre 2023). Malgré cette variation saisonnière, les concentrations restaient bien inférieures

aux normes nationales vietnamiennes pour les sédiments d'eau douce, correspondant à un niveau de pollution faible à modéré. La distribution des HAP en fonction des noyaux suivait l'ordre 4 cycles > 5 cycles ~ 3 cycles > 6 cycles > 2 cycles, avec Fluoranthène (Flt), Pyrène (Pyr), Benz[a]anthracène (B(a)A), Chrysène (Chr) comme HAP dominants, tandis que Nap et Ace étaient les moins fréquemment détectés. Les ratios diagnostiques indiquent que les HAP proviennent principalement de sources pyrogéniques, liées à la combustion de biomasse et de combustibles fossiles, reflétant l'influence des activités anthropiques telles que le trafic, l'agriculture et l'industrie. Les évaluations des risques (ERL/ERM, ISQG/PEL, TEQ/MEQ) suggèrent que les effets toxiques des HAP sur les organismes vivants et l'homme sont très faibles.

Le chapitre 4 présente une étude approfondie de trois familles de POPs chlorés ou bromés, à savoir les PCB, les PBDE et les CP, dans les sédiments du fleuve Rouge (Publication N°3). En évaluant leur occurrence, leurs variations spatio-saisonnières et les risques écologiques associés, ce chapitre apporte de nouvelles perspectives sur les problématiques de contamination liées aux produits chimiques industriels halogénés dans le fleuve Rouge.

Les PCB, importés au Vietnam dans la seconde moitié du XXe siècle et réglementés au niveau international depuis le début des années 2000, ont été détectés à de faibles concentrations (n.d. – 50,5 ng g^{-1}). La distribution spatiale et saisonnière de 7 congénères indicateurs suggérait des niveaux plus élevés pendant la saison sèche (novembre 2022) que pendant la saison des pluies (juin 2023). Ces faibles niveaux globaux ainsi que les dynamiques saisonnières sont en faveur d'un déclin progressif de la pollution par les PCB dans le fleuve indiquant une efficacité des mesures réglementaires nationales, mais cela n'a pas été clairement prouvé dans ce travail.

Les PBDE, utilisées commercialement plus tard que les PCB mais également réglementées à l'échelle internationale dans les années 2010, ont été détectées à de faibles concentrations (n.d. – 7,3 ng g^{-1}). Seuls les BDE 47 et 209 ont été systématiquement détectés dans les échantillons de sédiments de novembre 2022. Les données produites corroborent assez bien celles déjà publiées pour d'autres fleuves au Vietnam.

Les SCCP ont été observés à des concentrations nettement plus élevées (<LOD – 1515 ng g^{-1}), avec un schéma saisonnier marqué. Ces niveaux indiquent la pertinence croissante des CP en tant que contaminants émergents dans la rivière Rouge. Contrairement aux PCB, les CP ont montré des signes d'apports plus récents, avec des concentrations augmentant entre la saison sèche et la saison des pluies. L'augmentation temporaire des niveaux illustre les émissions continues provenant de diverses sources, probablement liées à leurs applications répandues dans les matériaux industriels, notamment dans le polychlorure de vinyle (PVC), qui a été produit en masse au Vietnam. Ce comportement met également en évidence leur mobilité dans les conditions hydrologiques du système du fleuve Rouge. L'augmentation suspectée des SCCP est préoccupante, car elle reflète des processus de contamination actifs plutôt que des résidus historiques de PCB.

L'évaluation des risques écologiques, réalisée selon plusieurs lignes directrices, a montré que la plupart des échantillons présentaient des risques faibles liés aux PCB et aux SCCP. Cependant, certains échantillons de la campagne de juin 2023 ont présenté des risques modérés associés aux SCCP. Si les émissions de CP se poursuivent, le risque écologique pourrait devenir plus significatif dans un avenir proche.

Le chapitre 5 présente une toute première évaluation de la contamination par les PFAS le long d'un transect de 200 km en aval du fleuve Rouge (Publication N°4). Cette étude a intégré à la fois des échantillons de sédiments et d'eau en combinant des approches ciblées et la méthode TOPA (Total Oxidizable Precursor Assay). Les concentrations en PFAS dans les sédiments variaient de <LOD à 1,5 ng g^{-1}, tandis que dans l'eau, elles variaient de 0,2 à 572 ng L^{-1}. La détection de multiples composés substances per- et polyfluoroalkylées (per- and polyfluoroalkyl substances, PFAS), incluant à la fois les acides perfluoroalkyliques (PFAA) et des alternatives émergentes telles que sulfonate de fluorotélomère 6:2 (6:2-FTS) et 6:2 di-perfluoroalkyl phosphatoéthanolamine (6:2-diPAP), reflète l'évolution des pratiques industrielles avec le remplacement de produits chimiques réglementés par des composés structurellement similaires. Ce type de substitution, qualifié de « regrettable replacement », souligne la difficulté de gérer ces contaminants émergents dont la persistance, le potentiel de bioaccumulation et les impacts écologiques ne sont pas encore pleinement compris.

L'application de la méthode TOPA a révélé une augmentation des niveaux et la formation de nouveaux PFAA non détectés par l'approche ciblée, mettant en évidence la contribution des précurseurs oxydables à la charge totale de PFAS dans les échantillons environnementaux. De plus, la différence d'équivalents fluor avant et après oxydation dans les échantillons d'eau et de sédiment a souligné les limites de l'analyse conventionnelle basée sur des composés ciblés. Des stratégies analytiques complémentaires, telles que l'analyse non ciblée, devraient être adoptées afin de détecter à la fois les molécules PFAS connues et inconnues.

Ce travail de thèse propose une évaluation originale de plusieurs classes de contaminants organiques dans le fleuve Rouge. Les résultats apportent des informations essentielles sur la dynamique saisonnière, les sources potentielles le long du fleuve et les implications réglementaires. Ces observations contribuent à l'appréciation des normes de qualité des sédiments, soutiennent le développement de stratégies de suivi et permettent une meilleure compréhension du panorama de la contamination organique dans ce système fluvial d'intérêt.

Mots clés : Vietnam, fleuve Rouge, golfe du Tonkin, HAP, PCB, PBDE, CP, PFAS, sédiments, eau.

Introduction

Vietnam is a developing country located in Southeast Asia. In the 20th century, the country had gone through several devastating wars. Beside the severe loss of human lives and the destruction of the nation's infrastructures, these wars had also induced detrimental and long-lasting damage on the Vietnamese environment. One of the most profound ecological consequences was due to the extensive use of chemical agents by the United States military. Notably, the military use of herbicides in Vietnam started in 1962 and peaked from 1967 to 1969. The herbicides (e.g., agent Pink, agent Orange, and agent Purple) were extensively sprayed during the Operation Ranch Hand by the US Air Force, as well as by ground troops and Navy riverboats along the riverbank (Institute of Medicine (US), 1994). The aim of this strategy was to facilitate the detection of the base camps of the opposing forces along the communication and infiltration routes as well as in the vicinity of the U.S. base camps, and to destroy the crops cultivated to supply for the opposing army. In particular, agent Orange, which is a mixture of 2,4-dichlorophenoxyacetic acid (2,4-D) and 2,4,5-trichlorophenoxyacetic acid (2,4,5-T), was the most widely used herbicides by US army in Vietnam during the war (Institute of Medicine (US), 1994, 2011). The Agent Orange was contaminated during the manufacture of 2,4,5-T by an unavoidable by-product, 2,3,4,7-tetrachlorodibenzo-p-dioxin (2,3,4,7-TCDD). 2,3,4,7-TCDD can interact with a gene regulatory protein, leading to an inappropriate modulation of gene expressions and causing a series of health effects in human (Pelclovä et al., 2006). During the period from 1961 to 1971, it has been estimated that the US military had released 74 million liters of tactical pesticides in Vietnam, of which 60% (43 million liters) were Agent Orange (Institute of Medicine (US), 2011). Young (2009), estimated that there would be 105-119 kg of dioxin in Agent Orange used in Vietnam. 4.8 million people, including civilians and military personals might have been exposed to the agent orange (Stellman et al., 2003). Vietnam's Ministry of Defense in 2012 estimated 3 million victims of the agent orange, the majority of which were the second and third generations of the exposed population (https://mod.gov.vn/vn/noi-dung/sa-ttk/sa-ttdv-tochuchoi/sa-ttk-ctch-hnncddc/sa-ttk-ctch-hnncddc-tl/6966f6b8-3398-

439d-a4df-fa2f3143e153, in Vietnamese, accessed July 2025). Despite the ongoing efforts by both the Vietnamese and US governments to remediate areas contaminated by Agent Orange, the long-term impacts on human health, future generations and the environment remain profound. While several clean-up projects are ended, like the dioxin remediation at Danang Airport was completed in 2018, dioxin continues to persist in soils and sediments, especially around former US military air bases in Bienhoa or Phucat. This has resulted in elevated rates of cancers and chronic illnesses among exposed local populations; and birth defects in the following generations.

Leaving behind wars and the irreversible damage caused by dioxin in Agent Orange, Vietnam entered into new era of development, especially after the economic reform "Đổi mới" in 1986. In 2024, the nation's GDP had increased by 7.1% (according to World Bank data, https://data.worldbank.org, accessed July 2025). Vietnam's population has surpassed 100 million. This development has led to constantly increasing consumer demand and rising purchasing power in the nation. To keep pace with this expanding domestic demand, the manufacturing sector has undergone rapid growth, while the importation of goods and services has increased considerably over the past few decades. The manufacturing industries contributed up to 26.3% of Vietnam's gross domestic product (GDP) in 2023 (Asian Development Bank, Asian Development Bank, https://kidb.adb.org, accessed July 2025). The total value of imports of goods and services to the country reached more than 300000 Millon US dollars. Besides the exceptional economic growth, Vietnam is now confronted with a range of environmental challenges, one of which is emission and discharge of chemical substances. The nation, similarly, to many other industrialized and industrializing countries, is now facing environmental risks associated with chemicals that are either used in industrial or agricultural processes or unintentional by-products. Organic pollutants can enter the environment through discharge points linked to industrial effluents, urban and agricultural runoff, or leachate from waste disposal sites, often as a results of insufficient treatment processes (Awonaike et al., 2022; Fuchte et al., 2022). Vietnam is now facing critical issues, concerning wastewater treatment. World Bank reported in 2013 that only 10% of domestic wastewater was treated in Vietnam (World Bank, 2013). According to the National Statistics Office (NSO), in 2015, only 5.3% of urban areas had wastewater treatment facilities that met the national standards (NSO,

https://www.nso.gov.vn/en/other-news/2025/01/the-status-of-wastewater-treatment-and-environmental-protection-in-vietnam-and-lessons-from-denmark/, accessed May 2025). The NSO further informed that many factories perform only preliminary wastewater treatment before discharging it into the environment. In such a context, one main hypothesis underlying the doctoral work, was that in Vietnam, a middle-lower income country, emissions of organic continants are largely under-documented, yet they are likely substantial in the context of rapid urbanization and industrialization and may evolve in the coming decades.

When entering the aquatic environment, organic pollutants can accumulate in sediment by particulate phase sedimentation, and through processes such as adsorption or partitioning, depending on their hydrophobicity. Sediment thus acts as a sink by trapping organic contaminants, some of which qualified as persistent or recalcitrant to degradation, will accumulate for decades, revealing sediment as a compartment less fluctuating than water and of long-term concern. However, certain conditions such as turbulence and flooding can cause remobilization of contaminants back to the water column, potentially impacting the water quality (Crawford et al., 2022a; Ducrocq et al., 2024). Therefore, sediment is not only sink but also secondary sources that strongly influence the occurrence of contaminants in the aquatic environment. As a consequence, it is crucial to perform regular sediment monitoring.

To tackle the issue of chemical pollution, on the international scale, the Stockholm Convention on Persistent Organic Pollutants (POPs) has been ratified by Vietnam since 2002. A series of National Technical Regulations have been further implemented by the Vietnamese government to define acceptable limits in environmental matrices, for various groups of hazardous chemicals, including persistent organic pollutants (POPs) and also heavy metals, polycyclic aromatic hydrocarbons (PAHs) or pesticides

Nonetheless, the existing regulatory framework only partially takes into account the whole range of molecules registered as POPs, and monitoring efforts are still limited for some recently including groups, specially chlorinated paraffins (CPs) or per- and polyfluoroalkyl substances (PFAS). Moreover, scheduled routine environmental monitoring is not yet comprehensive, leaving river basins and other vulnerable ecosystems insufficiently studied.

The Red River basin, one of Vietnam's most important waterways, stands as an example for these challenges. Passing through densely populated urban centers, rapidly industrializing zones, and intensively cultivated agricultural land, the Red River is subject to considerable anthropogenic pressures. Its strong seasonal hydrological fluctuations and high capacity of sediment transport further enhance the potential for the accumulation, remobilization, and redistribution of organic contaminants. Thus, the Red River, which flows from southern China through Hanoi, the densely populated capital, and other urban and industrial centers in the Red River delta, may act as a major transport pathway, conveying contaminants from inland areas to coastal zone.

According to the international and national regulatory context, to the urban, industrial and touristic development taking place in Vietnam and to the objective of tracking long-term pollution, three groups of pollutants were selected to document the quality status of the Red River. PAHs are ubiquitous organic contaminants that have been historically and extensively studied in the environmental compartments and can be nowadays considered as a scale for comparing the anthropization of worldwide environments. On the other hand, industrial chemicals under elimination by the Stockholm Convention over the last 20 years, including polychlorinated biphenyls, decabromodiphenyl ether (decaBDE) along with other polybrominated diphenyls (PBDEs), perfluorooctanoic acid (PFOA), perfluorohexane sulfonic acid (PFHxS), and short- or medium-chain chlorinated paraffins (CPs), present unresolved issues, as each of these groups consists of numerous congeners and molecules with different environmental behaviors, some of them being studied only since very recently. Unlike unintentional by-products such as the dioxins, mentioned earlier, which have historically received attention, particularly in the context of warfare in Vietnam, the later compounds are deliberately manufactured, extensively marketed, and present, throughout their diverse applications, several point-sources release into the environment, during production, storage, transport, usage, and end-of-life stage. Therefore, the doctoral work focuses on POPs in the Red River, spanning from well-documented compounds (i.e., PAHs) and legacy POPs (i.e., PCBs and PBDEs), to more contemporary and less studied groups gathering a huge number of congeners (i.e., CPs and PFAS), thus requiring more comprehensive screening approaches.

Based on this context, the doctoral research aimed at addressing the following scientific questions: What are the concentrations, spatial distributions, and seasonal

variations of PAHs, PCBs, PBDEs, CPs, and PFAS in the Red River? How do hydrological dynamics (dry and rainy season) and the different land use along the river influence the concentration levels and the fingerprint of these contaminants? How do the measured levels compare with Vietnamese regulations and international guideline values? And to what extent do current regulatory and monitoring framework sufficiently capture the environmental and health risks associated with both legacy and emerging contaminants?

To answer the scientific questions and test the proposed hypothesis, the general objective of my thesis is to contribute to the documentation and understanding of occurrence, spatial and seasonal distribution patterns, and risk assessment concerning the presence of PAHs, PCBs, PBDEs, CPs, and PFAS in Vietnamese aquatic environment. This will be tackled be a literature review and by three operational objectives aiming at addressing the gaps on the contamination situation of the Red River.

The thesis will focus on the ubiquitous PAHs, on PCBs representing historical POPs, whose production and use were banned decades ago, on PBDEs and CPs, which are considered as contemporary POPs, more recently included under Stockholm Convention, on PFAS, recognized as emerging POPs and many others currently under evaluation. The thesis will hence explore the dynamic relationship between regulatory efforts and the ever-changing variety of environmental contaminant emission.

The manuscript is organized into five chapters, which include four articles (one published, one accepted, and two to be submitted). The chapter one provides the international regulatory context of organic pollution and summarizes the knowledge on the contaminants of interest and reviewed the literature on their occurrence in Southeast Asian aquatic environment, in order to highlight the knowledge gaps underlying the thesis work and to precise the objectives of the research. In chapter two, the key methodological approaches and analytical techniques employed in this study are briefly overviewed and further detailed in the articles and Annex constituting each chapter's results. The results addressing the occurrence and distribution of PAHs, then PCBs, PBDEs, CPs, and finally PFAS in samples collected in the Red River, Vietnam are presented, as research articles, in chapters three, four, and five, respectively. The manuscript is ending with the main conclusions and perspectives.

1. Literature Review

Chemical pollution has become a major global environmental issue, driven by industrialization, urbanization, and the widespread use of synthetic compounds. Despite international efforts, the management of chemical contamination remains uneven, with developing regions often facing greater exposure and weaker regulatory frameworks. In Asia, rapid economic growth has intensified chemical use while monitoring and treatment infrastructures remain limited, making the region particularly vulnerable. Vietnam is also facing this challenge, where the combined effects of industrial expansion and limited chemical governance have increased pressure on aquatic ecosystems. Within this global and regional context, the management of chemical pollution has become a central challenge of environmental governance. International initiatives such as the Intergovernmental Science-Policy Panel on Chemicals, Waste, and Pollution (ISP-CWP) and the Stockholm Convention on persistent organic pollutants (POPs) aim to promote coordinated action to mitigate chemical risks. The latter has guided the selection of target compounds in this doctoral work, encompassing regulate and emerging contaminants. Chapter 1 introduces the international and regional frameworks for chemical management and presents the target contaminants of this study, covering both ubiquitous, historical, and emerging pollutants.

Understanding the occurrence and impacts of organic contaminants is based on an overview of the targeted contaminants and their chemical properties, sources, environmental behaviors, and regulatory frameworks. In chapter 1 an overview of groups of contaminants of interest is provided, beginning with their general chemical structures and fundamental physicochemical properties. In contrast with PAHs, which primarily originate from combustion processes and thus, are ubiquitous in the environment, the targeted POPs of industrial usage (i.e., PCBs, PBDEs, CPs, and PFAS) are presented in terms of their historical development, periods of widespread use, and commercial applications. esses and, thus, are ubiquitous in the environment.

Beyond their chemical and applications, the chapter examines how these substances enter the environment and human populations, focusing on exposure routes, environmental transport mechanisms, and biological interactions at the cellular level. Their documented toxicological effects, including carcinogenicity, endocrine disruption, and other chronic health outcomes, are also presented to illustrate their significance as organic pollutants.

The chapter reviews also the international and national regulatory frameworks. IN addition to the Stockholm Convention, which was intended to protect humans and the environment from their emissions, the chapter gives some insight on the regulations set by some globally recognized regulatory agencies such as United States Environmental Protection Agency or European Environment Agency, and Vietnamese national technical regulations to limit their occurrence and exposure doses.

The final section focuses the scope on the quality status of the aquatic environment in Southeast Asian countries and specifically in Vietnam, examining from the perspective of the dissemination of persistent organic substances. Thus, an overview of the knowledge on PAHs occurrence in Vietnam is summed up, followed by a broader examination of dissemination of PCBs, PBDEs, CPs, and PFAS across aquatic environments in Southeast Asia. This regional perspective highlights both the progress and challenges in addressing regulated and emerging pollutants in dynamic, yet vulnerable ecosystems such as the Red River, on which the doctoral work is focusing.

At the end of the chapter, gaps in knowledge and scientific questions raised by this literature review will be highlighted in order to explain in detail the objectives of the doctoral work and how to address them. Reviewing the available data and establishing the contextual framework are essential not only to introduce the environmental challenges taking place in Vietnam, but also to highlight the knowledge gaps that justify the orient the following chapters of the manuscript. By linking the findings of the first chapter to the structure of this thesis, the review illustrates how each chapter contributes to build a more complete quality status of the Red River.

1.1. Southeast Asia and Vietnamese context of development

Over the past decades, increasing awareness of the global threat posed by chemical pollution has led to the establishment of international conventions and frameworks aiming to reduce and prevent the release of hazardous substances into the environment. With these global efforts, one major milestone is the recent creation of the Intergovernmental Science-Policy Panel on Chemicals, Waste, and Pollution (ISP-CWP) by the United Nations Environment Program (UNEP). The panel seeks to strengthen the science-policy interface for managing chemical pollution and waste at the global scale (https://www.unep.org/isp-cwp, accessed in October 2025). This initiative recognizes that chemical contamination is a transboundary issue requiring coordinated international action.

In the context of global initiatives such as ISP-CWP, it is crucial to consider the regional dynamics of chemical consumption, release and persistence. One of ISP-CWP's objectives is to address regions with fragmented or limited knowledge about the extent of contaminant pollution, by addressing global data, identifying gaps, and supporting capacity development in under-studied areas. Consequently, Southeast Asia is a case in point. Southeast Asia, with a population of approximately 680 million, is undergoing rapid urbanization and economic expansion. The GDP growth rate reached 4.6% in mid-2024 according toAsian Development Bank, 2024). This rapid development, together with increasing urban populations (52.4% of southeast asian population live in urban area in 2024 , https://kidb.adb.org, accessed in October 2025), has led to higher consumption rates and a corresponding surge in importation of commercial goods to meet domestic and regional market demand (*Figure 1*). At the same time, the region has experienced an expansion in industrial sector, contributing between 22.7 to 60.7% of national GDP across Southeast Asian countries in 2024 (https://kidb.adb.org, accessed in October 2025), as well as a fast-growing tourism, which drives major infrastructure development and seasonal population pressure. Moreover, relocation of manufacturing industries from the Western countries has further increased production capacity, energy demand, and chemical use in sectors such as textiles, electronics, and plastics. Consequently, these activities amplify the pressures on natural resources and waste management systems.

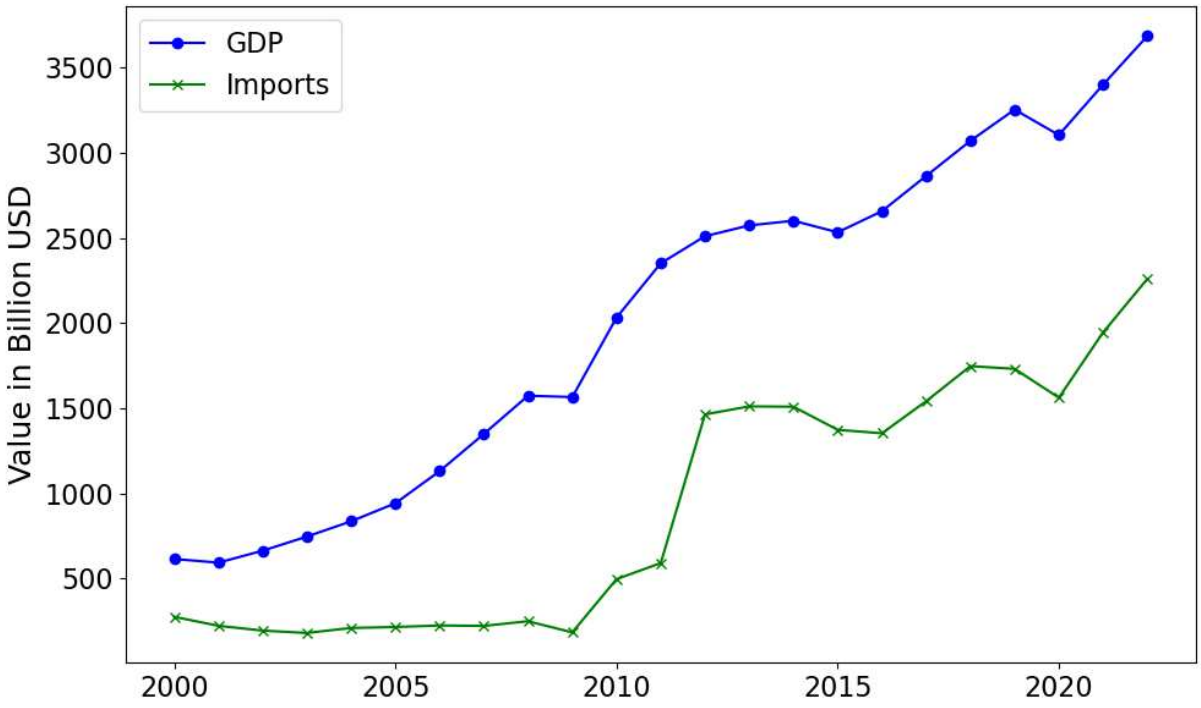


Figure 1: Increase in GDP (blue) and Imports (green) of Goods and Services in Southeast Asia between 2000 and 2022, data obtained from https://kidb.adb.org, accessed in October 2025.

Within this regional picture, Vietnam represents a particularly relevant case of examining the occurrence and dynamics of chemical contaminants. Over the past two decades, the country has experienced remarkable economic growth and industrial expansion, becoming one of the fastest-growing economies in Southeast Asia with GDP's growth rate of 7.1% in 2024. This development, while contributing to social and economic progress, has also intensified environmental pressures, particularly in urbanized and instrialized river basins such as the Red River Delta.

Vietnam, like many Southeast Asian countries, is located within the intertropical zone, where ecosystems are exposed to high temperatures, intense humidity, and strong hydrological variability. These climatic conditions, combined with rapid urbanization, can accelerate the degradation, transformation, and remobilization of organic contaminants. Additionally, the region's high vulnerability to climate change by frequent extreme events such as typhoons, floods, and prolonged drought, may significantly alter the transport pathways and distribution of pollutants. The Red River, with its strong seasonal regime, provides a representative case study of how tropical rivers respond to environmental

pressure and how climate-driven phenomenon can influence contaminant behavior and occurrence.

The Red River originates in southern China and flows through northern Vietnam, passing through Hanoi, the densely populated capital of Vietnam, as well as industrial and agricultural areas of the Red River Delta, befor discharging into the Gulf of Tonkin. The combination of industrial discharge, domestic wastewater, agricultural runoff, and rapid urbanization has made this river a hotspot for anthropogenic pressures. Despite the country's commitment to environmental protection, environmental surveys on the occurrence of novel entities such as synthetic chemicals remain limited in the river system. Understanding their occurrence is, hence, essential not only for assesing local environment quality but also for contributing to the regional and global objectives promoted by initiatives like the ISP-CWP. Such knowledge will provide a scientifc foundation for Vietnam's ongoing efforts to strengthen chemical management and mitigate the possible associated risks (e.g. the new Chemical Law in 2025 includes stricter regulations on imports, enhances safety management throughout the chemical lifecycle, and focuses on green technology).

1.2. International frameworks

The international framework for managing chemical pollution is founded on a set of evolving conventions that combine principles of environmental transparency with concrete measures to control persistent and/or hazardous contaminants. The Aarhus Convention (United Nations Economic Commission for Europe, UNECE, 2010) establishes the public's right to access environmental information, participate in environmental decision-making, and seek justice in environmental matters, thereby promoting transparency and accountability in pollution control. Building on this foundation, the Aarhus Protocol on Persistent Organic Pollutants (POPs), adopted under the Convention on Long-range Transboundary Air Pollution (CLRTAP), was designed to reduce or eliminate emissions of persistent, bio-accumulative, and toxic substances that pose transboundary risks (UNECE, 2010). Among the substances targeted by the Protocol, polycyclic aromatic hydrocarbons (PAHs) are listed to be reduced under the emission obligation. Under the terms, the member countries commit to reduce the emissions of PAHs to levels below those of 1990 (or a year between 1985 and 1995). The European Union has put a maximum allowable concentration of 270 ng L^{-1} for BaP in inland surface

waters (Directive 2013/39/EU). While the standard limits of 11 out of 16 priority PAHs are well established for freshwater and marine sediments (QCVN 43:2017/BTNMT), there is no established regulation or maximum limit for PAHs in surface waters set by the Vietnamese government.

Expanding this regulatory approach to a global scale, the Stockholm Convention on POPs was adopted in 2001, to protect human health and the environment from chemicals presenting four main characteristics: persistence in the environment (e.g. half-life in soil of DDT is 2 to 15 years and that of PCB is 1 to 5 years, Blaylock, 2005; Terzaghi et al., 2021), bio-accumulate through the food web, and pose risks to ecosystems and humans, and can be transported far from their primary source of emission.

As of 2025, the Stockholm Convention has 186 Parties, and 152 of which are signatories. The convention has achieved near-universal participation, showing global commitment to control and eliminate POPs.

The Southeast Asian countries have signed and ratified the Stockholm convention since its early implementation stage. The date of signatures and ratification by these countries are presented in the *Table 1.*

Table 1: Status of ratification of the Stockholm Convention in Southeast Asian Countries

Southeast Asian countries	Signature	Ratification	Entry into force
Brunei Darussalam	21/05/2002	-	-
Cambodia	23/05/2001	25/08/2006	23/11/2006
Indonesia	23/05/2001	28/09/2009	27/12/2009
Laos	05/03/2002	28/06/2006	26/09/2006
Malaysia	16/05/2002	-	-
Myanmar	-	19/04/2004	18/07/2004
The Philippines	23/05/2001	27/02/2004	27/05/2004
Singapore	23/05/2001	24/05/2005	22/08/2005
Thailand	22/05/2002	31/01/2005	01/05/2005
Timor-Leste	-	-	-
Vietnam	23/05/2001	22/07/2002	17/05/2004

Under the Stockholm Convention, Parties commit to eliminate or restrict the production, use, and release of listed POPs (Annexes A and B) and to minimize unintentional emissions (Annex C). They must also ensure the environmentally sound management of POP wastes and contaminated stockpiles and submit National Implementation Plans to the Convention. However, the level of implementation varies among countries, depending on their technical capacities and the dates of ratification. Consequently, not all Parties are at the same stage in monitoring or controlling these compounds in environmental matrices, which partly explained the differences observed in the availability of data on POPs in the environment.

The Convention initially listed twelve POPs consisting mainly of pesticides such as DDT, aldrin, and dieldrin, alongside a few industrial chemicals like PCBs and unintentional by-products (i.e., dioxins and furans). Over time, the list of regulated substances has increased, indicating new scientific evidence and the emergence of contaminants of concern (*Table 2*). Notably, a growing number of industrial compounds have been added, many of which exist as complex mixtures or families of congeners.

Table 2: The POPs listed in the Stockholm Convention.

	POPs	Year of inclusion	Remarks
Annex A - Elimination	**Aldrin**	2001	Pesticide
	Chlordane	2001	Pesticide
	Chlordecone	2009	Pesticide
	Chlorpyrifos	2025	Pesticide
	Decabromodiphenyl ether	2017	Industrial chemical
	Dechlorane Plus	2023	Industrial chemical
	Dicofol	2019	Pesticide
	Dieldrin	2001	Pesticide
	Endrin	2001	Pesticide
	Heptachlor	2001	Pesticide
	Hexabromobiphenyl	2009	Industrial chemical
	Hexabromocyclododecane	2013	Industrial chemical

	Hexa- and heptabromodiphenyl ethers	2009	Industrial chemical
	Hexachlorobenzene	2001	Pesticide, Industrial chemical
	Hexachlorobutadiene	2015	Industrial chemical
	Alpha hexachlorocychlochexane	2009	Pesticide
	Beta hexachlorocyclohexane	2009	Pesticide
	Lindane	2009	Pesticide
	Medium-chain chlorinated paraffins	2005	Industrial chemical
	Methoxychlor	2023	Pesticide
	Mirex	2001	Pesticide
	Pentachlorobenzene	2009	Pesticide, Industrial chemical
	Pentachlorophenol and its salts and esters	2015	Pesticide
	Polychlorinated biphenyls	2001	Industrial chemical
	Polychlorinated naphthalenes	2015	Industrial chemical
	Perfluorooctanoic acid, its salts and related compounds	2019	Industrial chemical
	Perfluorohexane sulfonic acid, its salts and related compounds	2022	Industrial chemical
	Short-chain chlorinated paraffins	2017	Industrial chemical
	Technical endosulfan and its related isomers	2011	Pesticide
	Tetrabromodiphenyl ether and pentabromodephenyl ether	2009	Industrial chemical
	Toxaphene	2001	Pesticide
	UV-328	2023	Industrial chemical

Annex B - Restriction	**DDT**	2001	Pesticide
	Perfluorohexane sulfonic acid, its salts and related compounds	2009	Pesticide, Industrial chemical
Annex C – Unintentional production	Hexachlorobenzene	2004	Unintentional Production
	Hexachlorobutadiene	2017	
	Pentachlorobenzene	2009	
	Polychlorinated biphenyls	2001	
	Polychlorinated dibenzo-*p*-dioxins	2001	
	Polychlorinated dibenzofurans	2001	
	Polychlorinated naphthalenes	2015	

As an active member state of the Convention since 2002, Vietnam has made efforts to identify and inventory the presence of POPs nationwide and has accordingly implemented additional legal frameworks to meet the Stockholm Convention's objectives. For instance, in 2018, the Vietnamese government has amended the Law on Chemicals by issuing degree 82/2022/ND-CP which restricts the industrial use of several POPs, i.e., polychlorinated biphenyls (PCBs), decabromodipheyl ether (decaBDE) along with other polybrominated diphenyls (PBDEs), perfluorooctanoic acid (PFOA), perfluorohexane sulfonic acid (PFHxS) and short-chain chlorinated paraffins (SCCPs).

Therefore, the present work focuses on intentionally-produced industrial POPs that include polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs), chlorinated paraffins (CPs), and poly- and perfluoroalkyl substances (PFAS). These substance have been widely used in industrial applications and exhibit high chemical stability, strong carbon-halogen bonds, and a remarkable resistance to environmental degradation (ATSDR, 2000, 2017; Evich et al., 2022; Muir et al., 2000). While PCBs represent historical POPs, whose production and use were banned decades ago, PBDEs and CPs are considered as contemporary POPs, more recently included under Stockholm Convention. Notably, medium-chain CPs (MCCPs) have officially been listed as POPs in

2025. Moreover, PFAS, a diverse chemical class, are now recognized as emerging POPs with some molecules (e.g., PFOS, PFOA, and PFHxS) already listed and many others currently under evaluation (e.g., perfluoroalkyl acids with carbon chains C_9 – C_{21}) (UNEP, 2022a).

1.3. Introduction to contaminants of interest

The contaminants on which the doctoral work is focused, represent a wide spectrum of environmental contaminants, ranging from ubiquitous (PAHs), to historical to historical (PCBs), hardly degradable pollutants, with decades of monitoring efforts, to contemporary and emerging POPs of industrial use (PBDEs, CPs, and PFAS), which continue to challenge analytical chemistry and regulations. The inclusion timeline in the Stockholm Convention of POPs of interest was summarized in *Figure 2*.

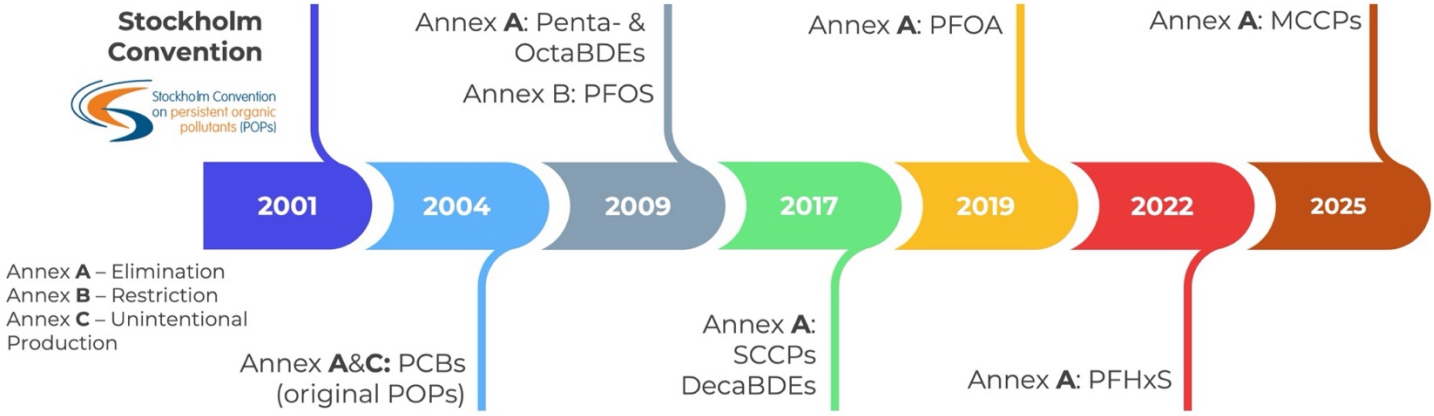


Figure 2: The inclusion timeline of POPs of interest in the Stockholm Convention

These compounds illustrate the dynamic and adaptive nature of international chemical regulation, which evolves in response to adverse in experimental monitoring and growing awareness of global contaminant dispersion. The following sections discuss their structure, sources, and environmental occurrence, presented by chemical family

1.3.1. Structures, synthesis, and physicochemical properties

1.3.1.1. Polycyclic Aromatic Hydrocarbons

Polycyclic Aromatic Hydrocarbons (PAHs) are organic compounds that contain at least two fused benzene rings. The *Figure 3* shows the chemical structure of Chrysene, a PAH with 4 benzene rings. PAHs originate from both natural and anthropogenic sources. Anthropogenic activities represent the dominant sources of PAHs in the environment, including fossil fuel and biomass combustion, petroleum cracking, production of carbon-based and petroleum-derived materials, emission from vehicle engines and waste incineration, and oil spillage (Giuliani et al., 2008; Saha et al., 2009). In addition, natural processes such as volcanic eruptions and forest fires can also contribute to PAH emissions.

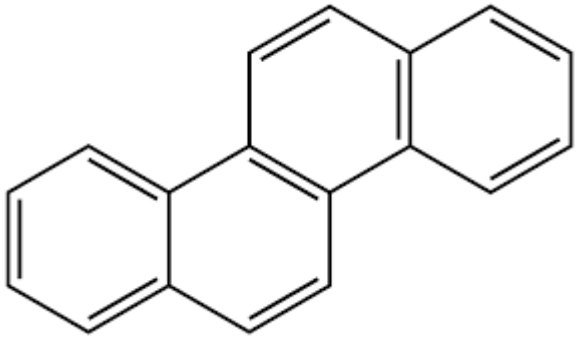


Figure 3: Chemical structure of Chrysene

Among the hundreds of PAHs present in the environment, the United States Environmental Protection Agency (US-EPA) has identified 16 priority PAHs (*Annex 1*) since 1970s due their toxicity. (ATSDR, 1995). According to their molecular weights, 16 PAHs can be classified into 2 groups: low molecular weights (LMW) PAHs consisting of 2 and 3 benzene rings and high molecular weights (HMW) with 4-6 benzene rings.

1.3.1.2. Polychlorinated Biphenyls

Polychlorinated biphenyls (PCBs) are a group of synthetic chemicals consisting of a biphenyl structure bound to hydrogen and chlorine atoms. PCBs can be produced by the chlorination of two benzenes (biphenyls) with anhydrous chlorine under the catalytic

condition (with the presence of ferric chloride, $FeCl_3$, ATSDR, 2000; Ngoubeyou et al., 2022). The results are congeners with general formula of $C_{12}H_{10-x-y}Cl_{x+y}$. The general structure of PCBs is shown in *Figure 4.*

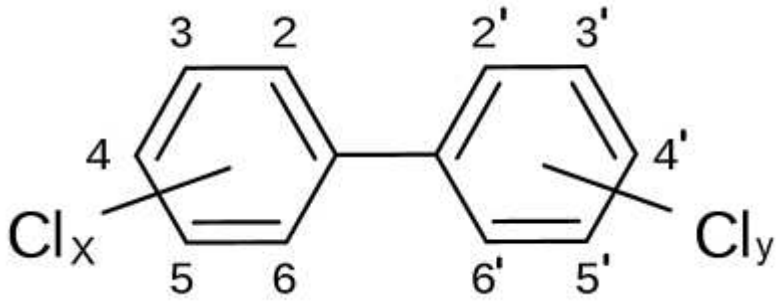


Figure 4: General structure of Polychlorinated biphenyls (obtained from Passatore et al., 2014)

Based on the number of chlorine atoms substituting the hydrogen ones and their position on the biphenyl structure, the International Union of Pure and Applied Chemistry (IUPAC) has recognized 209 congeners of PCBs, numbered from PCB – 1 to PCB – 209. According to the degree of chlorination, twelve homologues were identified: monochloro- (3 congeners), dichloro- (12), trichloro- (24), tetrachloro- (42), pentachloro- (46), hexachloro- (42), heptachloro- (24), octachloro- (12), nonachloro- (3), and decachlorobiphenyl (1).

The characteristics of individual PCB congeners largely depend on their level chlorination. They exhibit a range of physcial properties, from colorless, oily, highly mobile liquids to more viscous and darker substances and yellow to black resins (Borja et al., 2005). PCBs have outstanding dielectric and electrical insulating properties, along with non-flammability and strong resistance to chemical and thermal degradation. (Vergani et al., 2017; Zhu et al., 2021).

1.3.1.3. Polybrominated diphenyl ethers

Polybrominated diphenyl ethers (PBDEs) are a group of organic chemicals that consists of a diphenyl ether structure with bromine atoms substituting hydrogen atoms of the benzene rings. The general chemical formula is $C_{12}H_{(10-n)}Br_nO$. The general chemical structure of PBDEs is shown in *Figure 5*.

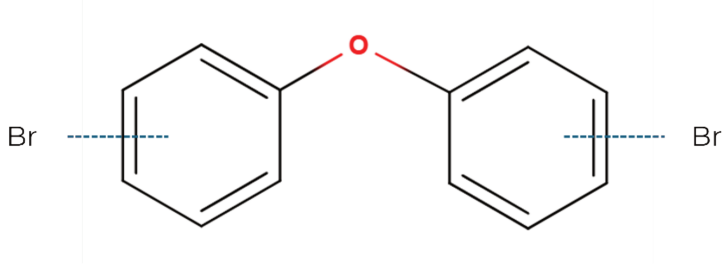


Figure 5: General structure of PBDEs

Similarly to PCBs, depending on the numbers of bromine atoms and their position on the diphenyl structure, IUPAC has identified 209 distinct congeners and categorized them into 10 different groups from mono- to decabromodiphenyl ethers.

PBDEs are synthesized by the bromination of diphenyl ethers under catalyzed conditions, resulting in mixtures of brominated diphenyl ethers (Akortia et al., 2016; Darnerud et al., 2001). Due to the presence of an oxygen atom and the steric hindrance of PBDEs, only a limited number of congeners are formed (Alaee et al., 2003). Their formation generally follows a sequential pathway, beginning with PBDE 47, which serves as a precursor for BDE 99 and BDE 100; these subsequently give rise to BDE 153 and 154, followed by BDE 183 and utimately BDE 209. Depending on the degree of bromination, PBDEs exhibit wide variations in physicochemical properties. As the number of bromine atoms increases, volatility and water solubility decrease, while their physical state shifts from viscous liquids in lower-brominated congeners to solids in highly brominated congeners (Akortia et al., 2016; Alaee et al., 2003).

1.3.1.4. Chlorinated Paraffins

Chlorinated paraffins (CPs) are mixtures of polychlorinated alkanes with varying degrees of chlorination. The chlorine mass fraction in CPs ranges from 30 to 70% weight/weight (w/w). The general chemical formula of CPs is $C_nH_{2n+2-x}Cl_x$. An example of CPs is illustrated in *Figure 6.*

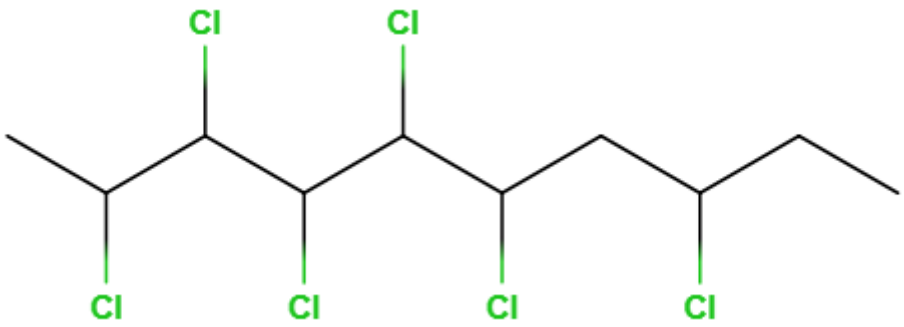


Figure 6: C_{10} chloroparaffin, 61% Cl by weight

CPs are produced through the radical chlorination of n-alkanes derived from petroleum distillation under high pressure and/or in the presence of ultraviolet radiation, which initiates the substitution of hydrogen atoms by chlorine (van Mourik et al., 2015). Due to the low selectivity of the reaction, the process yields complex mixtures containing thousands of constitutional isomers, diastereoisomers, and enantiomers.

According to the length of their carbon chains, CPs are divided into three major groups

- Short-chain chlorinated paraffins (SCCPs) refer to CPs containing between 10 and 13 carbon atoms.
- Medium-chain chlorinated paraffins (MCCPs) refer to CPs containing between 14 and 17 carbon atoms
- Long-chain chlorinated paraffins (LCCPs) refer to CPs containing 18 and more carbon atoms.

Recent literature has also proposed additional categories, including very short-chain chlorinated paraffins (vSCCPs, C_6-C_9) and very long-chain chlorinated paraffins (vLCCPs containing more than 21 carbon atoms, Ding et al., 2021; Wu et al., 2022; Y. Zhou et al., 2019).

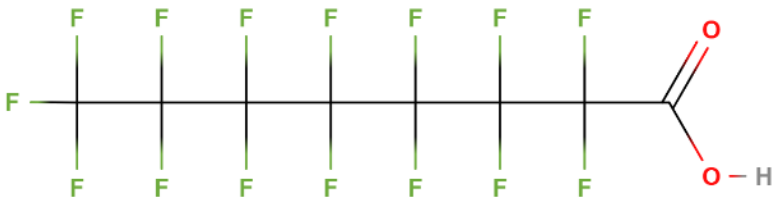
CPs are viscous and colorless mixtures or yellowish dense oils, although solid forms can be obtained when the chlorine content is high (70%, typically in CPs with carbon chain lengths of C_{20} – C_{30}, Muir et al., 2000). CPs are characterized by their strong hydrophobicity with Kow values ranging from around 5 to 13 (H. Wang et al., 2019) . Their volatility and solubility in water generally decrease as the number of carbon and chlorine atoms increases with the exception of CPs with fewer than 6 chlorine atoms showing semi-volatile behavior (Muir et al., 2000), or certain CPs having some degree of molecular

polarity depending on both number and spatial arrangement of chlorine atoms (Glüge et al., 2013).

1.3.1.5. Poly and Per-fluoroalkyl Substances

Per- and Polyfluoroalkyl substances (PFAS) are fluorinated organic compounds that contain at least one fully fluorinated methyl or methylene carbon atom (without any H/Cl/Br/I atom attached to it, OCED, 2021). Perfluoroalkyl substances are aliphatic compounds whose hydrogen atoms bonded to carbon atoms are all replaced by fluorine atoms, whereas polyfluoroalkyl substances have only some of their hydrogen atoms replaced by fluorine. Two examples of per- and polyfluorinated compounds are shown in *Figure 7a* and *Figure 7b*, respectively.

A



B

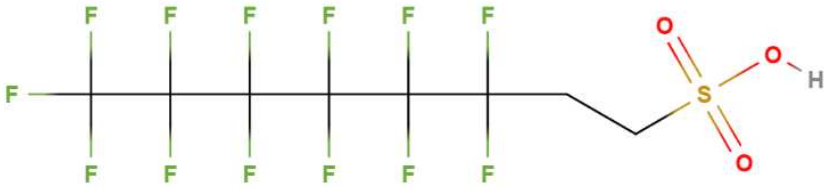


Figure 7: The chemical structure of a) perfluorooctanoic acid (PFOA, a perfluorinated compound) and b) 6:2-fluorotelomer sulfonic acid (6:2-FTS, a polyfluorinated compound)

It is estimated that over 8000 chemicals of known structures have been identified as PFAS (Evich et al., 2022). PFAS can be divided into two major groups: non-polymeric PFAS and polymeric PFAS (*Figure 8*). Non-polymeric groups consists of perfluoroalkyl and polyfluoroalkyl substances. Polymeric PFAS comprise of fluoropolymers, perfluoropolyethers, and side-chain fluorinated polymers.

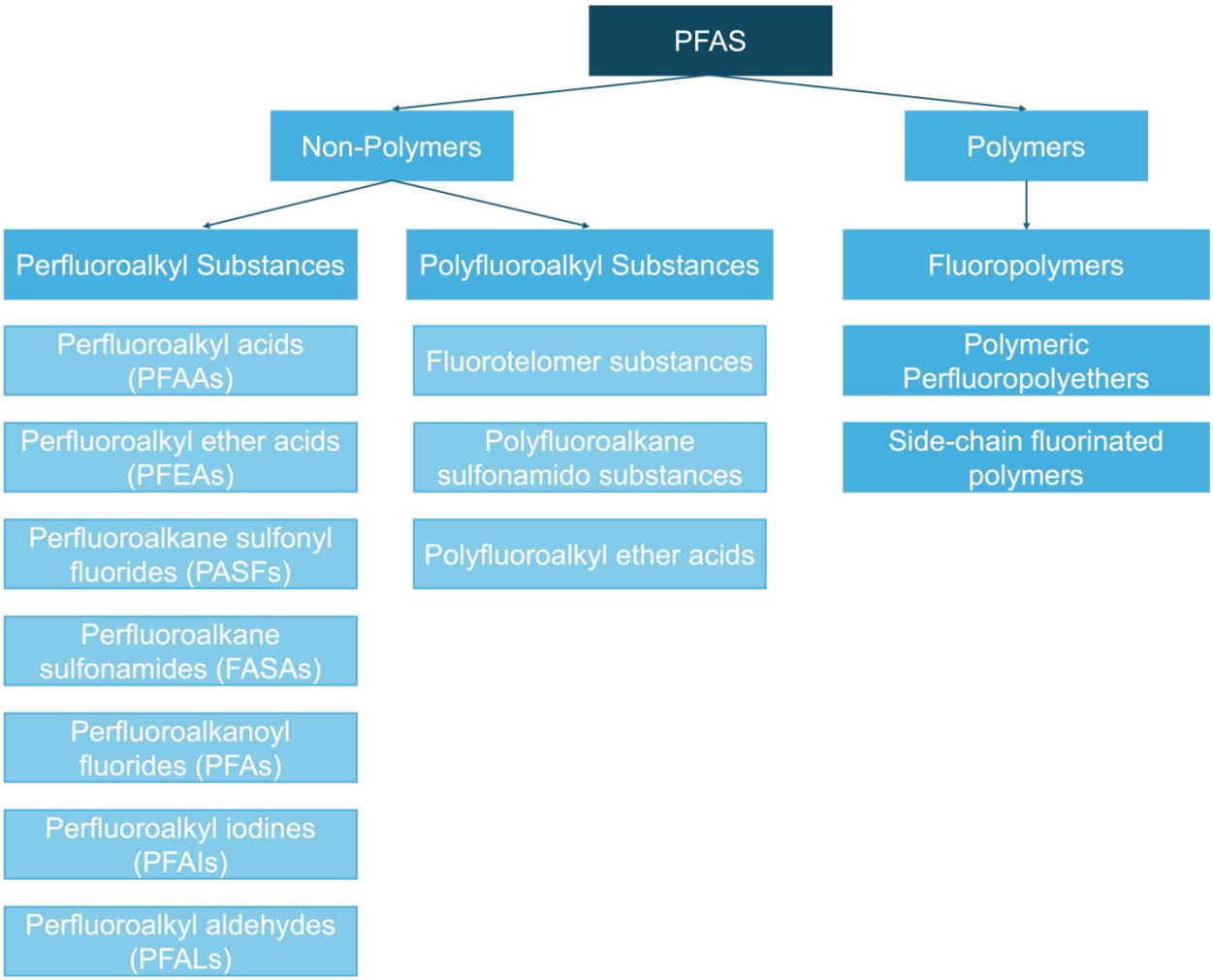


Figure 8: PFAS familly, adapted from (Buck et al., 2021; J. Li et al., 2022)

According to Leung et al. (2023), PFAS typically consist of a perfluorinated alkyl chain coupled with a hydrophilic head group. The fluorinated alkyl tail is highly hydrophobic and oleophobic, largely due to the nature of the C-F bond, which is otherwise the strongest single bond in organic chemistry. The bond provide both a steric barrier and an electrostatic shield around the carbon backbone, as fluorine atoms carry significant electron density and create strong dipolar resonance structures along the chain. Such structural features reduce chemical reactivity, resist thermal decomposition, and limit biological and chemical degradation processes.

1.3.2. Production, emission sources, and environmental behaviors.

1.3.2.1. PAHs

PAHs can be classified into two groups based on their origin. The first groups, petrogenic PAHs, forms through the low-temperature transformation of organic matter and is predominantly composed of LMW compounds. These PAHs are mainly associated with crude oi and petroleum products. The second group, pyrogenic PAHs, is generated during the incomplete combustion of fossil fuels and biomass at high temperatures. Pyrogenic PAHs are generally HMW compounds (Kishida et al., 2007; Pradhap et al., 2021).

The environmental emissions of PAHs arise from multiple pathways: residential and commercial biomass burning (firewood, coal, crop residues), open biomass burning (wildfires, agricultural residue burning), fossil fuel combustion (gasoline, coal, oil), industrial combustion, and waste burning. PAHs can be emitted as both particulate-bound and gaseous forms, with fine particulate matter (e.g., PM2.5) as a carrier (Z. Li et al., 2023), enabling long-range atmospheric transport and deposition onto both terrestrial and aquatic ecosystems. A comprehensive global inventory (Shen et al., 2013) estimated that in 2007, the total atmospheric emission of 16 priority PAHs was approximately 504 gigagrams per year (Gg yr^{-1}). The major contributing sectors were residential and commercial biomass burning accounting more than 60% of the total emissions, open-field biomass burning (13.6% of total emissions) and petroleum consumption by motor vehicles (12.8% of total emissions). The authors also carried out scenarios modelling and reported that PAH emissions peaked at around 592 Gg in 1995 and gradually declined to about 499 Gg in 2008.

In aquatic environments, PAHs enter through direct discharges from industrial effluents, urban runoff, and atmospheric deposition. Once in water bodies, due to their low water solubility, their hydrophobic nature, and strong affinity for organic matter, PAHs tend to bind to suspended particles and accumulate in sediments, leading to their accumulation in bottom sediments, and potential bio-accumulation in aquatic organisms and bio-magnification along the trophic webs, posing potential long-term ecological and human health risks (Vijayanand et al., 2023). Degradation of PAHs in aquatic systems is

influenced by factors such as temperature, microbial activity, and the presence of oxygen (Berríos-Rolón et al., 2025), but is generally slow, implying their persistence in the environment for long-term.

1.3.2.2. PCBs

Being first synthesized by Oskar Gustav Doebner, a German chemist in 1876, PCBs had been commercialized since the 1920s (Ngoubeyou et al., 2022). PCBs were primarily produced commericially in the US from 1929 untill 1977 with globally marketed trade names such as Aroclor, Askarel and Therminol. Commerial PCB mixtures also have different trade names depending on the place of manufacture, e.g., Clophen (Germany), Fenclor (Italy), Kanechlor (Japan), and Phenoclor (France). It was reported that approximately 1 to 1.5 million tons of PCBs have been produced since the 1920s, equivalent to 17 million tons of PCBs-containing products (Othman et al., 2022). PCBs were historically produced in many countries, for examples, more than 640,000 tons in the United States (US), over 170,000 tons in former Soviet Union, around 134,000 in France, over 58,000 tons in Japan and approximately 10,000 tons in China (Breivik et al., 2002; H. Yu et al., 2023).

PCBs were extensively utilized in electrical and electronic equipment, as heat transfer fluids in cooling systems and hydraulic machinery, and as plasticizers in paints, adhesives, sealants, and rubber products (De & Pereira, 2004; Tue et al., 2010). Despite their ban, PCBs still remain in transformers and capacitors which are either in use or in storage, especially in China and Southeast Asian countries (Vu et al., 2025).

Given their wide applications and mass production, PCBs have gradually entered into the environment and have been detected in atmospheric particles and dust (H. Q. Anh et al., 2020; Tue et al., 2010), soil (Qin et al., 2022; Thao, Cau, et al., 1993), water and sediments (Mbusnum et al., 2020; Unyimadu & Benson, 2023). Reddy et al., 2019 proposed numerous contamination pathways for PCBs, including waste discharge from the production sites, leakage from dumping sites of electronic wastes, and evaporation from paints and coatings. Due to their hydrophobicity, PCBs have a tendency to accumulate in atmospheric and aquatic particulate matter, giving them the capacity for long-range transport. PCBs have been found in remote regions, where there is no sign of neither human nor industrial activities (Eckhardt et al., 2007). More dangerously, the

lipophilic nature of PCBs allows them to bind with the lipid segments in organism tissues (Reddy et al., 2019). There has been evidence of their presence in organisms and human.

1.3.2.3. PBDEs

Commercial PBDE formulations are typically categorized into three main mixtures: pentaBDE, octaBDE, and decaBDE, which differ in bromine content and congener composition (Kefeni et al., 2011). The PentaBDE mixture mainly contains tetraBDEs (24 – 38%), pentaBDEs (50 – 62%), and a small proprotions of hexaBDE (4 – 8%) (ATSDR, 2017). The congener composition of commonly used octaBDE mixtures (i.e., DE-79 and Bromkal 79-8DE) was determined by La Guardia et al. (2006) While DE-79 mixture consisted of approximately 10.5% hexaBDEs, 45% heptaBDEs, 38% OctaBDEs and over 13% of nona- and deca-BDEs, Bromkal 79-8DE contained more than 33% hepta- and octaBDEs, 19% nonaBDEs and over 49% BDE 209. The authors also pointed out that BDE 209 was the major congeners for DecaBDE mixtures (over 90%) with nonaBDEs accounting for arround 5%.

The penta-BDE mixture was predominantly used as a flame retardant in polyurethane foams, particularly in furniture and mattresses (Costa & Giordano, 2014; Renzelli et al., 2023). OctaBDE and decaBDE mixtures were primarily incorporated into various industrial and consumer products, including plastics, textiles, and electronic devices such as computers, televisions, and household appliances (Costa & Giordano, 2014; Renzelli et al., 2023). Octa-BDE mixtures were used mainly in acrylonitrile-butadiene-styrene polymers, usually at 12-18% by weight whereas 90% of decaBDE were incoporated in plastic used in electrical and electronic equipment (Jinhui et al., 2017).

From 1970s until 2005, 1.1 to 1.5 million tons of PBDEs have been globally produced, more than 70% of which is decaBDE (T. Meng et al., 2021; Motamedi et al., 2022).

The production and the consumption of PBDEs reached their peak during the late 1990s and early 2000s. Abbasi et al. (2019) estimated that the production of PBDE peaked at 85,000 tons yr^{-1} in 2003 (*Figure 9*). In the same year, the consumption of penta- and decaBDE peaked at 12,000 tons yr^{-1} and 75,000 tons yr^{-1}, respectively. In the early 2000s, PBDE manufacturing gradually shifted from the United States to Asian, where China, having initiated PBDE production in the 1980s, appeared as one of the major global produceres of DecaBDE by 2000.

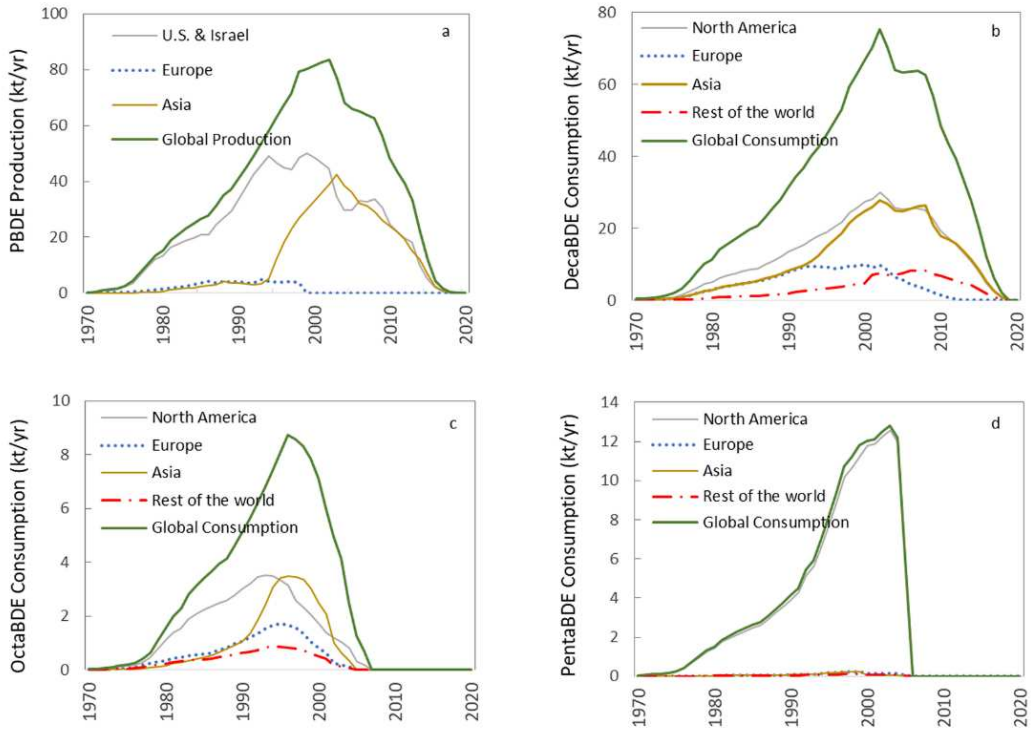


Figure 9: The estimated a) production of PBDEs and consumption of b) DecaBDE mixture, c) OctaBDE mixture, and d) PentaBDE mixture from 1990 to 2020, figure obtained from Abbasi et al. (2019).

Given that PBDEs are physically added to the final products rather than chemically bound, they can leach and escape the plastic matrix and enter the surrounding environment (Costa & Giordano, 2014; Darnerud et al., 2001). As a consequence, this persistent group of chemicals has been found in dust (Hoang, Tran, et al., 2021; X. Yu, Liu, et al., 2022), water (Q. Chen et al., 2024; Olaniyan et al., 2024), sediment (Salvadó et al., 2012; Toan et al., 2020) , biota (Falahudin et al., 2022; D. T. Nguyen et al., 2024), and human (Ermler & Kortenkamp, 2022; J. Zhang et al., 2017).

1.3.2.4. CPs

The production of CPs began in the 1903s and until 1977, CPs were produced exclusively in the US, with the annual production from 20,000 to 37,000 tons yr^{-1} (Glüge et al., 2016). After 1977, production expanded to Europe and Japan. By the 2000s, China had become the world's major producer, exceeding 1,000,000 tons yr^{-1} in 2013. Until today, large-scale production continues primarily in China and India (C. Chen et al., 2022). Published reports furthermore indicated that China, the world's largest producer and

exporter of SCCPs, produced nearly 200,000 tons between 2021 and 2022 (K. Fang et al., 2025). C. Chen et al. (2022) estimated that 32.5 million tons of CPs were produced between 1930 and 2020, with 28% being SCCPs, 57% MCCPs and 15% LCCPs.

Being produced since 1930s, CPs have been widely used for their versatile properties. Their main applications include extreme pressure additives in metal working fluids, plasticizers in polyvinyl chloride (PVC) production, and flame retardants in plastics, paints, rubbers, leathers and sealants (Bayen et al., 2006; McGrath et al., 2023).

With wide application in the industrial processes, CPs have raised concern about their presence in the environmental matrices i.e., air, dust, water, soil, sediment and biota, especially aquatic species (L. Chen et al., 2022; South et al., 2022; Tahir et al., 2024; Wu et al., 2020).

According to the global emission estimation be C. Chen et al. (2022), 5.2 million tons of CPs were released into the environment between 1930 and 2020, with annual emissions peaking at 193,000 tons yr^{-1}. SCCPs and LCCPs each contributed about 30% of total historical emissions, while MCCPs accounted for 40% (*Figure 10a*). Soils and freshwater were identified as the primary environmental sinks (*Figure 10b*). Moreover, wastewater treatment plants receiving industrial and municipal effluents from CP production and use were major emission sources to the environment (*Figure 10c*).

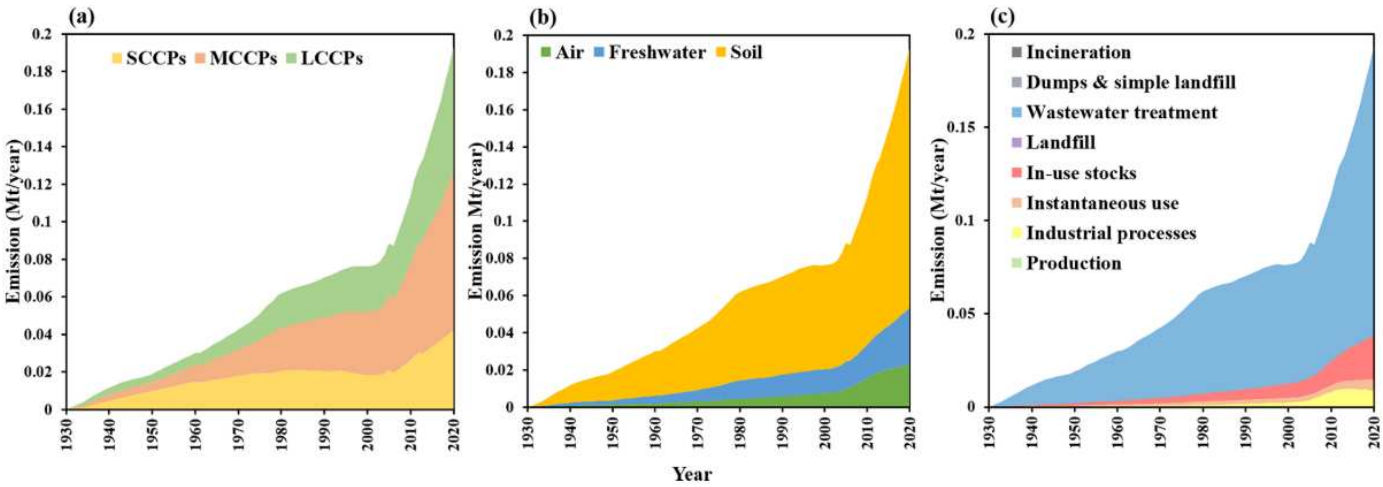


Figure 10: Global missions from 1930 to 2020 of a) SCCPs, MCCPs, and LCCPs to b) different receiving environment from c) different emission sources . Figure obtained from (C. Chen et al., 2022).

1.3.2.5. PFAS

Since the mid-20th century, large-scale industrial production of FPAS expanded rapidly. Between 1951 and 2004, the global production of PFCAs, mainly ammonium pefluorootanoate (APFO, the ammonium salt of PFOA) and ammonium perfluorononaoate (APFN, the ammonium salt of PFNA), was estimated at 4400 to 8000 tons, with the principal manufacturing facilities located in the US and Belgium (Prevedouros et al., 2005). Similarly, the production of perfluorooctane sulfonyl fluorid (POSF), a key prescursor for PFOS, was estimated at approximately 96,000 tons. 3M company was the major global manufacturer at that time, producing 78% of total global POSF production in 2002. Following the voluntary phase-out of POSF by 3M, global PFAS production underwent major shift from the US and Europe to Asia, particularly China (Z. Wang et al., 2014). Industrial-scale manufacturing in China began around the year 2000, with initial production volumes estimated at less than 50 tons yr^{-1} before 2003. However, as the Western producers stopped the production in response to regulatory and environmental concerns, China expanded its PFAS industry, reaching the annual production of 120 tons yr^{-1} by 2012 (C. Xu et al., 2025). The annual production of PFAS in China had gradually decreased from 251 tons in 2015 down to 120 tons in 2022 (Cai et al., 2025).

Since the 1940s, PFAS have been extensively used in commercial and industrial applications, including firefighting foams, water-repellent and waterproof products, lubricants, food packaging and processing equipment, non-stick cookware, textiles, and furniture (Dickman & Aga, 2022; Glüge et al., 2020; Herzke et al., 2012).

Given their large-scale production, long-term use, and diverse industrial applications, PFAS have been released into the environment throughout their production and usage. Global emission inventories indicated that between 26,100 and 21,400 tons of C_4 – C_{14} PFCAs were emitted between 1951 and 2002, respectively, with fluoropolymer manufacturing as the dominant sources, contributing around 55 to 83% of total historical PFCA releases (Wang et al., 2014). China in 2014 became the primary global emitter, with an estimated 80 tons of PFOA released in 2014 (Song et al., 2018).

Cumulative emissions over the period 1958-2015 have been reported at 1229 to 4930 tons for PFOS and 1230 to 8738 tons for PFOS-related precursors (Z. Wang et al., 2017). Other sulfonic acid homologues have also contributed substantially to global PFAS emissions. For instance, total emissions of PFHxS and perfluorodecan sulfonic acid (PFDS)

between 1958 and 2015 were estimated from 120 to 1022 tons and from 38 to 378 tons, respectively (Boucher et al., 2018). More recently, cumulative emissions from China between 2015 and 2022 reached 467 tons, highlighting the country's continued dominance in global PFAS production and consumption.

Because of their vast applications, PFAS have become ubiquitous in the environmental settings (Evich et al., 2022; Z. Li et al., 2024; Zhong et al., 2021). PFAS are often referred to as "forever chemicals" due to their strong carbon-fluorine bonds, among the strongest in chemistry, which make them resistant to degradation (OCED, 2021; K. Singh et al., 2023a).

PFAS exhibit a strong potential for bioaccumulation in aquatic and terrestrial organisms due to their chemical stability and resistance to metabolic degradation (Burkhard & Votava, 2023; Macorps et al., 2022; Murakami et al., 2011). These compounds have been shown to be biomagnifying through successive trophic levels within the food web, leading to progressively higher concentrations in higher consumer species (Boisvert et al., 2019; George et al., 2023). Consequently, PFAS can be detected in a wide range of food items, including fish, meat, dairy products, and crops (T. Bonato et al., 2025).

1.3.3. Hazardous effects

1.3.3.1. PAHs

PAHs are among groups of organic pollutants that have been extensively investigated due to their widespread environmental distribution and their well-documented toxicological effects on human health. Chronic exposure to PAHs has been proven to series of adverse health effects in human including cancers, damage to DNA, immune and reproductive systems, neurological impairments and developmental disorders (ATSDR, 1995). Benzo(a)Pyrene (BaP) is, among all PAHs, considered the most extensively studied carcinogen for humans. BaP metabolites have been shown to form adducts within human DNA, inducing the mutations of *p53* gene and causing lung cancer (Alexandrov et al., 2002). (Perera et al., 2005) assessed the DNA damage by measuring BaP-DNA adducts in mothers and newborns. They found that adduct levels were similar or even higher in infants compared to their mothers, suggesting that newborns may be more susceptible to DNA damage. This heightened vulnerability could increase the risk of developmental disorders and cancers later in life. Not only in mothers and babies, but DNA adducts have

also been detected in sperm cells (Gaspari et al., 2003). The presence of these adducts can compromise the integrity of parental genome, potentially leading to infertility.

1.3.3.2. PCBs

Several PCB congener can exhibit toxic effects similar to those of polychlorinated dibenzodioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs). Such congeners, consisting of four or more chlorine atoms with one or no substation at the ortho positions, are called dioxin-like PCBs (Reddy et al., 2019). The World Health Organization (WHO) has listed 12 PCBs (PCB 77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169 and 189) as dioxin-like and established toxic equivalency factors (TEFs) for risk assessment (Van den Berg et al., 2006).

PCBs are considered as endocrine disrupting chemicals and responsible for several disorders and toxic effects in humans, including cancers, disruption of reproductive systems, developmental and immunological problems, and metabolic and thyroid diseases. (Buha Djordjevic et al., 2020). Having the similar structure to thyroid hormone, the PCBs' hydroxylated metabolites can disturb the thyroid receptors, acting as both agonist and antagonist (Zani et al., 2019). DeCastro et al. (2006) pointed out 13 congeners having estrogenic activities due to their chlorination pattern associated with the estrogen affinity. Furthermore, a fetus can be exposed to PCBs through the umbilical cord during pregnancy, while newborns and children may be exposed through breast milk and food, potentially leading to impaired cognitive and motor development (El Majidi 2014).

1.3.3.3. PBDEs

When entering human bodies, PBDEs have been proven to be hazardous. The metabolites of PBDEs, having the similar structure to thyroid hormone, have high affinity to thyroxin transport proteins, thus suppressing the normal binding mechanism of thyroid hormones (Renzelli et al., 2023). Metabolites of BDE-47 was proven to interfere the binding of Tetraiodothyronine (T4- thyroid hormone, Gao et al., 2024). Sheikh & Beg (2020) even suggested the higher affinity of BDE-99's metabolites to disrupt the transport of thyroid hormone when compared to those of BDE-47. Moreover, PBDEs may also induce several toxicities, including hepatoxicity, neurotoxicity, genotoxicity and developmental toxicity, and immunotoxicity (Costa & Giordano, 2014; Couderc et al., 2016; Gao et al., 2024).

1.3.3.4. CPs

Alarmingly, there has been considerable evidence of the presence of CPs, especially SCCPs, in human organs and body fluids, e.g., hair, nails, blood, breast milk, placenta tissues and umbilical cord blood (J. W. Huang et al., 2023; Mu et al., 2023). A study carried out by Krätschmer et al. in 2021 on breast milk samples collected from 53 countries concluded that CPs constituted from 18 to 46% of the total POPs concentration in the samples, much higher than PCBs or PBDEs.

Human exposure to CPs can occur through multiple pathways, with the primary routes being dietary intake, inhalation of contaminated air and dust, and dermal contact (van Mourik et al., 2016). Occupational exposure is particularly relevant for individuals working in the CPs manufacturing industries or in recycling and treatment sites for CPs-containing materials (Mu et al., 2023). In vivo experiments have shown that CPs, especially SCCPs, can disrupt thyroid hormone balances, cause developmental abnormalities, and lead to impairments in the immune and neurological systems (Darnerud & Bergman, 2022; J. W. Huang et al., 2023). CPs can also be carcinogenic and SCCPs were categorized in Group 2B as possibly carcinogenic to human by International Agency for Research on Cancer (IARC, https://monographs.iarc.who.int/list-of-classifications, assessed March 2025).

1.3.3.5. PFAS

Human exposure to PFAS primarily occurs through the consumptions of contaminated water and food. This exposure is somehow driven by the definition of what is a PFAS (e.g. trifluoroacetic acid is very concentrated in the dissolve water phase and is considered a PFAS only since the 2021 OECD definition). Reports suggested the presence of PFAS in drinking water, bottled water, animal products and vegetables (T. Bonato et al., 2025; Guardian et al., 2020; Kunacheva et al., 2010). Inhalation and dermal absorption are also recognized as potential exposure routes, given the presence of PFAS in materials such as waterproof paints, textiles, carpets, and other household furnishings (Glüge et al., 2020; Herzke et al., 2012). In the human body, due to strong affinity to serum albumin, PFAS have a tendency to partition to and accumulate in protein-rich organs (Starnes et al., 2024). PFAS have been found in internal human organs, including tissues in liver, kidney, lung and brain, placenta, human embryos and fetal organs (Domingo, 2025; Mamsen et al., 2019). PFAS have been reported to be responsible for causing cancers, disturbances in endocrine,

immune, neurological and reproductive systems, and several other disorders (Blake & Fenton, 2020; Fenton et al., 2021).

1.4. Environmental occurrence of PAHs, PCBs, PBDEs, CPs, and PFAS in Southeast Asian aquatic environment

1.4.1. PAHs occurrence in Vietnamese aquatic environment

The first study on sedimentary PAHs in Vietnam was conducted by M. T. Anh et al. in 1999. The sediment samples were collected from urban and sub-urban canals and rivers in Ho Chi Minh City, the largest city in Vietnam. The author reported the concentration of Σ9 PAHs ranging from 67.5 to almost 15000 ng g^{-1} dw. The highest level of PAHs was detected in sample collected in an urban canal, which received wastewater from nearby industrial factories. A summary of the PAH occurrence in sediment samples collected in Vietnam is presented in *Table 3.*

Table 3: Comparison of PAH concentrations (ng g^{-1}) in sediment samples across Vietnam

Type of samples	Number of PAHs	Concentration range (ng g^{-1} dw)	Reference
Sediments from urban and sub-urban canals and rivers in Ho Chi Minh City (n = 10)	8	67.5 - 14947	M. T. Anh et al. (1999)
Sediment from Kim Nguu River, center of Hanoi (n = 5)	16	1820 - 5120	Boll et al. (2008)
Sediment from coastal lagoons in central Vietnam (n =9)	14	95 - 465	Giuliani et al. (2008)
Urban canals and Saigon River in Ho Chi Minh City (n = 9)	14	256 – 4617	Saha et al. (2009)

Sediment from Nhue and To Lich River (n = 5)	15	84.4 - 2766	Duong et al. (2014)
Sediment the Cau River in Bac Giang Province (n = 24)	15	27 - 3949	Giuliani et al. (2019)
Sediment from Saigon River (n = 11)	16	49 – 933	Babut et al. (2019)
Estuarine sediment in Cangio district (n = 18)	15	0.73 – 518	(Thuy et al., 2021)

A series of studies was carried out in Hanoi, the capital city of the country, and the northern region in the late 2000s and 2010s. Boll et al. (2008) analyzed 16 PAHs in sediment samples collected in the urban river of Kim Nguu. Concentrations across five sampling sites ranged from 1820 to 6210 ng g^{-1} dw. Similar levels of PAHs were also reported in sediment samples from other urban rivers in Hanoi, including the Nhue and To Lich Rivers, where the concentrations of 15 priority PAHs were from 84.4 to 2766 ng g^{-1} dw (Duong et al., 2014). Comparable levels were found in the Cau River, located in Bac Giang province, with the concentration of 15 PAHs from 27 to almost 4000 ng g^{-1} dw (Giuliani et al., 2019). Additionally, considerable attention has been given to Ho Chi Minh City, a major metropolitan area in the southern Vietnam, and the broader Mekong Delta region. Sediment samples collected from urban canals (n = 6) and the Saigon River (n = 3) in Ho Chi Minh City showed elevated concentrations of 14 PAHs, ranging from 551 to 1888 ng g^{-1} dw in canals, and from 256 to 4617 ng g^{-1} dw in the river (Saha et al., 2009). However, more recent studies reported slightly lower levels of PAHs in the Saigon River. For example, Babut et al. (2019) measured concentrations of 16 PAHs in the Saigon River from 49 to 933 ng g^{-1} dw. Similarly, 15 PAHs in sediment from the estuarine zone of Can Gio district, downstream of the Saigon River and predominantly rural area, could reach up to 518 ng g^{-1} dw (Thuy et al., 2021). In the Vietnamese central coastal area, 16 priority PAHs could also be detected in sediment samples from nine lagoons, varying from 95 to 465 ng g^{-1} dw (Giuliani et al., 2008). The majority of studies have shown that, in the sediment, medium-to-high molecular weight PAHs (e.g., Phe, Pyr, and BaA) were the predominant congeners. These compounds are typically associated with pyrogenic sources.

Due to PAHs' tendency to accumulate in the suspended matter and sediment, a few articles reported the occurrence of these contaminants in the dissolve water compartment. Trinh et al. (2017) analyzed 21 PAHs in water samples collected in paddy fields before and after flooding events in the central Vietnamese provinces of Hue and Thanh Hoa. The results show an increase in PAHs levels following the floods, with concentration rising by 1.5 to 2 times in both provinces. The surge in PAHs levels highlighted a significant risk of contamination to water resources in flooded areas. Another research working on PAHs in water was conducted by (Salingay et al., 2024), in which passive samplers were deployed for at least 30 days at 3 sites along Can Tho River, located in the heart of the Mekong Delta. The total concentrations of 16 PAHs ranged from 9.1 to 35.6 ng L^{-1}. Nap and 3-ring PAHs were the most dominant, accounting for more than 87% of the total PAHs across sampling sites.

1.4.2. Occurrence and levels of PCBs, PBDEs, CPs, and PFAS in water resources of eight southeast Asian countries

This section is the modified version of the article "Occurrence and levels of industrial persistent organic pollutants under the Stockholm Convention: A review of water resources in Southeast Asian countries" (Vu et al., 2025) published in August 2025 in Journal of Hazardous Materials Advances. It aims to review the available data on the occurrence of PCBs, PBDEs, CPs, and PFAS in aquatic compartments (i.e., water, sediment, and aquatic species) in Vietnam and, more broadly, across eight Southeast Asian countries. It also examines how international regulations, particularly those under the Stockholm Convention, have influenced the environmental contamination levels of these chemicals. The article is the continuation of section 1.4.1. on PAHs in order to achieve the first objective of the doctoral work, involving at reviewing the present knowledge on the four groups of persistent pollutants of industrial usage in Southeast Asia, in view of highlighting beneficial and/or novel research directions to undertake.

Publication N°1: **Occurrence and Levels of Industrial Persistent Organic Pollutants under the Stockholm Convention: A Review of Water Resources in Southeast Asian Countries**

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Occurrence and levels of industrial persistent organic pollutants under the Stockholm convention: A review of water resources in southeast Asian countries

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ABSTRACT

The Stockholm Convention (SC) on Persistent Organic Pollutants (POPs) aims to protect the environment and human health by regulating the production and the use of the so-called forever chemicals. Among the compounds listed under the SC, are 209 congeners of Polychlorinated biphenyls (PCBs) and Polybrominated biphenyl ethers (PBDEs), Chlorinated paraffins (CPs) and three molecules and related compounds of Poly- and Perfluoroalkyl substances (PFAS), representing four groups of industrial POPs gathering a huge number of molecules. These chemicals have been extensively used in various applications since the mid-20th century, resulting in their global dispersion as persistent and long-range-transport substances. Moreover, the rapid industrialization and economic development in Southeast Asian countries have heightened the vulnerability of their environments to such pollutants. The differences, both in the phases of economic development and in the history of practices and rulings, implies specific trends of POP concentrations in these developing countries. This articles reviews in sections three to five the occurrence and the reported concentrations of PCBs, PBDEs, and PFAS in the fresh and marine hydro-systems, gathering samples of water, sediment, and aquatic biota, studied across eight Southeast Asian countries over the past three decades. The summarized data provides in-depth information for the national policymakers to align with the objectives of the SC. It shows that these countries are paying increasing attention to document the quality status of the water resources, particularly in connection with the implementation of regulations, or with the edible resources. But the efforts need to be strengthened. Indeed, the review underscores in section 6, the urgent need for the regular environmental monitoring of these broad groups of POPs, points out the knowledge gaps in some countries and for some POPs, and highlights future directions for research to ensure the sustainability of water resources.

Graphical abstract





Abstract

The Stockholm Convention (SC) on Persistent Organic Pollutants (POPs) aims to protect the environment and human health by regulating the production and the use of the so-called forever chemicals. Among the compounds listed under the SC, are 209 congeners of Polychlorinated biphenyls (PCBs) and Polybrominated biphenyl ethers (PBDEs), Chlorinated paraffins (CPs) and three molecules and related compounds of Poly- and Perfluoroalkyl substances (PFAS), representing four groups of industrial POPs gathering a huge number of molecules. These chemicals have been extensively used in various applications since the mid-20th century, resulting in their global dispersion as persistent and long-range-transport substances. Moreover, the rapid industrialization and economic development in Southeast Asian countries have heightened the vulnerability of their environments to such pollutants. The differences, both in the phases of economic development and in the history of practices and rulings, implies specific trends of POP concentrations in these developing countries. This articles reviews in sections three to five the occurrence and the reported concentrations of PCBs, PBDEs, and PFAS in the fresh and marine hydro-systems, gathering samples of water, sediment, and aquatic biota, studied across eight Southeast Asian countries over the past three decades. The summarized data provides in-depth information for the national policymakers to align with the objectives of the SC. It shows that these countries are paying increasing attention to document the quality status of the water resources, particularly in connection with the implementation of regulations, or with the edible resources. But the efforts need to be strengthened. Indeed, the review underscores in section 6, the urgent need for the regular environmental monitoring of these broad groups of POPs, points out the knowledge gaps in some countries and for some POPs, and highlights future directions for research to ensure the sustainability of water resources.

Key words: PCBs, PBDEs, PFAS, CPs, Southeast Asia, aquatic environment, water resources.

1. Introduction

Southeast Asia (SEA) is located in the Western Pacific region, and consists of 11 countries (Brunei, Cambodia, Indonesia, Laos, Malaysia, Myanmar, the Philippines, Singapore, Timor Leste, Thailand, and Vietnam). With a population of 680 million, of

which 51.8% are urban population (2023, Asian Development Bank), the region has been witnessing a high rate of urbanization and rapid growth in the economy (GDP growth rate at 4.6% in July 2024, Asian Development Bank, 2024). The economic development and the increase in urban population lead to higher consumption, prompting greater importation of commercial products in order to meet the population's need and that of the expanding markets. In addition to importation, the manufacturing industries in Southeast Asia are rapidly expanding in the response to the consumers' demand. The manufacturing industries accounted for 9.2 – 24.9% of national GDP in 2023 in the region (Asian Development Bank, https://kidb.adb.org, accessed November 2024). Naturally, the increase in production rate leads to higher demand for chemicals used for various industrial purposes. The environment in SEA is thus vulnerable to anthropogenic contaminants, given the rise in importation and fast development in the industrial sectors in the region. To protect the environment, along with their own national regulations, Southeast Asian countries have either signed or ratified the Stockholm Convention (*Annex 5*) with the commitment to regulate the use of Persistent Organic Pollutants (POPs) and eliminate their discharge to the environment.

POPs are groups of organic substances with specific physical and chemical properties so that they i) remain highly persistent to degradation, ii) have the capacity for long-range transport in the environment and can iii) accumulate and become toxic in living organisms and human beings. The Annex D of the Stockholm Convention has defined the four criteria when identifying POPs (UNEP, 2023b). To be considered persistent, a chemical's half-life must be greater than two months in water and six months in soils. A chemical is regarded as bio-accumulative if its bio-concentration factor or bio-accumulation factor in aquatic species is greater than 5000, or its Kow is greater than 5. The potential for long-range environmental transport refers to the measured levels of the chemical in locations distant from the sources of its release. The transport may occur via air, water or migratory species. And the toxicity includes adverse effects to human health and to the environment. There were, initially, 12 POPs in the Stockholm Convention, nine of which were pesticides, e.g. Aldrin, Chlordane, and Hexachlorobenzene. Throughout the year, more and more organic chemicals have been listed in the Convention, making the total number of 34 POPs (https://www.pops.int/TheConvention/ThePOPs/AllPOPs, assessed November 2024) identified as pesticides (20 POPs) and industrial (18 POPs) or unintentional chemicals (7 POPs). These 34 POPs are classified into three Annexes: Annex

A (the production and use of chemicals must be eliminated), Annex B (the production and use of chemicals must be restricted), and Annex C (the unintentional releases of chemicals must be reduced). The majority of POPs are listed as individual contaminants. Among the industrial POPs, a few stand out as they are not just individual compounds but rather consist of a large number of molecules. This is in particular the case for the polychlorinated biphenyls (PCBs) and the polybrominated diphenyl ethers (PBDEs) gathering 209 congeners each; Chlorinated paraffins (CPs) and the three poly- and perfluoroalkyl substances (PFAS), their salts and related compounds, representing several thousands of molecules. If the occurrence, levels and distribution of these POPs were widely described in Europe, America and Japan or China, it is needed to zoom in countries, where the implementation of the Convention is difficult or where there is limited capacity to apply it. Indeed, the differences, both in the phases of economic development and in the history of practices and rulings, mean that the trends of concentrations of POPs measured in developed part of the world cannot be extrapolated to the developing countries (Mbusnum et al., 2020). According to the inventory reports from the Southeast Asian nations, even though there has been no recorded production of these four industrial POPs, they are still present in large quantity in the region due to the contaminated products and materials. It was estimated that there were approximately 30,000 tons of oil containing PCBs imported to Vietnam from the USSR, China and Romania (Kishida et al., 2007). The accumulative amount of PBDE impregnated in vehicles imported in Thailand was estimated to be 1.5 tons (UNEP, 2022b). And several countries in the region have imported several tons of PFAS annually in form of firefighting foams and synthetic materials (UNEP, 2014, 2017, 2022b).

POPs could be released into the environment from both point (e.g., production, usage, and waste disposal sites) and non-point sources (e.g., runoff and wastewater discharge, atmospheric deposition sources); and may distribute unevenly across environmental compartments, influenced by their physicochemical properties. PCBs, PBDEs and PFAS have been detected in fresh and marine surface water and sediment (Kwan et al., 2014; Lam et al., 2017; Wang & Kelly, 2017), ground- and drinking water (Hongkachok et al., 2017, 2023), and biota from low to high trophic levels (Falahudin et al., 2023; Yudhistira Azis et al., 2021). Water is regarded as a transport vector for contaminants. Industrial wastewater, urban and agricultural runoff, or leachate from waste disposal sites can introduce organic chemicals into the environment via discharge points, often stemming

from inadequate treatment efficiency (Guardian et al., 2020; Lertassavakorn et al., 2021). Besides water, sediment has been considered a sink and a source of POPs in the aquatic environment. On the one hand, the sediment is increasingly considered a sink accumulating contaminants through various sorption mechanisms related to chemical, physical and biological processes (Lampert, 2018). This phenomenon can be further enhanced by the dam-induced trapping of sediment. Dam construction has been proven to increase the retention time of sediment in the reservoirs, thus enhancing the accumulation and pollutant loads (J. Guo et al., 2023). Ang et al. (2024) reported that there were 1055 dams in the Mekong River basin, designating the region as a potential storage hotspot for POPs and more broadly, organic contaminants. On the other hand, sediment is subjected to disturbance caused by extreme events and bioturbation. In this case, the sediment becomes a source releasing the contaminants back into the water column. Moreover, sediment is a crucial component in the hydro-system, hosting numerous species, e.g., benthic invertebrates. These invertebrates could take up and accumulate the anthropogenic contaminants that are gradually biomagnified in the food chains, along the trophic levels (Bayen et al., 2005; Brandsma et al., 2015). The Southeast Asian countries are well connected via several river systems (e.g., Mekong River, Irrawaddy River and Chao Phraya River) and marine waterways (e.g., East Sea, Strait of Malacca, Gulf of Thailand and Celebes Sea). Given the long-range-transport capability of POPs, the transboundary contamination among Southeast Asian countries is possible. As other actual sources, PCBs, PBDEs or PFAS may be vectored by or released from microplastics (Bouhroum et al., 2019; Yeo et al., 2020; F. Yu et al., 2024), the presence of which has been highly reported in recent years in Southeast Asian countries (Curren et al., 2021; Gabisa et al., 2022; T. T. Nguyen et al., 2024). Extreme weather events (e.g., flooding, seas level rise, and typhoons) driven by climate change impact the remobilization and redistribution of contaminants in the environments (Bolan et al., 2024; Crawford et al., 2022b). Southeast Asia is one of the most vulnerable regions to climate change and expected to be exposed to more severe rainfall and higher sea level rise by the middle of the 21st century (Intergovernmental Panel on Climate Change (IPCC), 2023). Therefore, it is necessary to investigate the presence of POPs in Southeast Asian aquatic compartments to better evaluate their potential sources and fate in the ecosystem.

For the last decade, a few review articles on PCBs, PBDEs and PFAS in this region were published. These reviews either generally covered emerging POPs in selected matrices

(Bhagat et al., 2024) or focused on specific groups of contaminants at smaller geographical scales (Kaw & Kannan, 2017; Mackintosh et al., 2015). Hence, the present work aims to comprehensively review the scientific discoveries on the contamination of the aquatic environment, i.e. water, sediment, and biota samples, by PCBs, PBDEs, CPs, and PFAS in the 11 Southeast Asian countries, that are hydro-geographically connected and therefore relevant to study together.

This review highlights also research gaps and suggests future directions to make progress in the study of POP analysis and dispersion, particularly in the context of development and climate change in Southeast Asia.

2. Methodology

This review covers the occurrence of four groups of industrial POPs namely PCBs, PBDE, CPs, and PFAS in three aquatic matrices, i.e., in sediment, waters (fresh and marine surface water, ground water, drinking water) and aquatic biota in SEA. "Occurrence", "environment", "contamination", the names of the groups of organic contaminants and the name of each Southeast Asian country (*Figure 10 and Annex 5*) were the keywords used as "Web of Science" queries. The review only took into account the research articles and papers written in English. The preliminary results, totaling 1642 items, revealed the following geographical distribution: 9 articles in Brunei, 27 in Cambodia, 152 in Indonesia, 92 in Laos, 318 in Malaysia, 7 in Myanmar, 96 in the Philippines, 396 in Singapore, 228 in Thailand and 317 in Vietnam. A more comprehensive search was necessary to identify articles that investigated at least one group of target compounds in at least one relevant environmental matrix (i.e., water, sediment, and biota) within one or more of the 11 Southeast Asian countries. To filter the results, the authors carefully reviewed the title and abstract of each article. Articles were excluded if they did not study any of the contaminants of interest, focused on unrelated topics (e.g., politics, social welfare) or covered outside the target geographic region. For example, because the acronym "PCBs" is also used to refer to "Printed circuit boards", several studies related to electronics were initially included in the search results but were later excluded during screening. After this comprehensive search, a smaller set of relevant articles remained. Finally, 6 articles were reported in Cambodia, 17 in Indonesia, 3 in Laos, 14 in Malaysia, 15 in the Philippines, 20 in Singapore, 23 in Thailand and 61 in Vietnam (*Figure 11*). No articles were found regarding Brunei, Myanmar, and Timor-Leste. Moreover, no articles

were found investigating the occurrence of CPs in the region in the environmental matrices of interest. As a result, this article reviews the occurrence and levels of three groups of POPs, namely PCBs, PBDEs and PFAS, in eight Southeast Asian countries (i.e., Cambodia, Indonesia, Laos, Malaysia, Philippines, Singapore, Thailand and Vietnam).

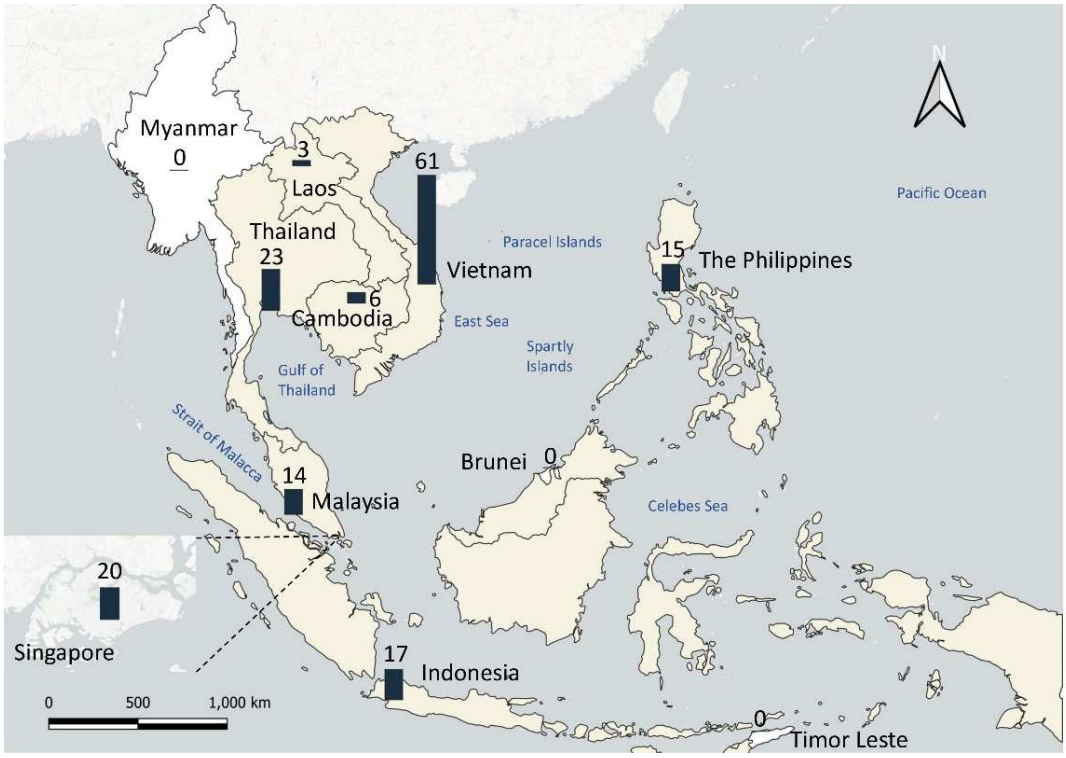

Figure 11: Map of Southeast Asian countries covered by the review, and for each country the number of articles published from 1990 to 2024 and dealing with at least one of the three studied groups of POPs

The total number of 121 articles (as reported in Annex) were reviewed and compiled (*Annex 6*), covering 35 years from 1990 to the 1st of April 2024. The trend of the number of articles published per year on PCB, PBDE, PFAS occurrence, in Southeast Asia, over the period, is presented on *Figure 12*, which clearly shows a real beginning of the research studies from 2003 (enter in force of the Stockholm Convention) and an average of 8 papers per year since. The sum of articles in individual countries *(Figure 11)* was higher than the sum which can be calculated from *Figure 12* because one article might work on samples from multiple countries and could be counted several times. Sixteen review

articles were published during this period and excluded from our review, but the data, if it matched our focus, was retrieved from the primary publication.

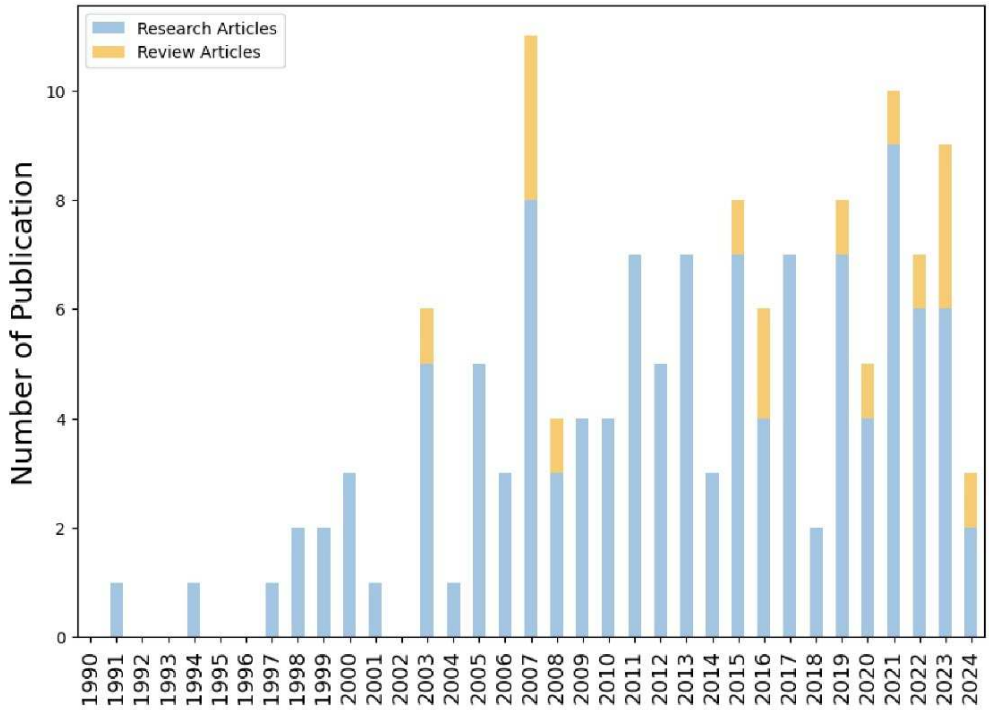

Figure 12: Number of publications reporting PCBs, BPDEs, and/or PFAS concentrations in water, sediment, and aquatic biota samples in eight Southeast Asian countries from 1990 to 2024

All the data compiled in the present review are summarized in *Annex 6*. The data used for the graphical presentations were the average values of the reported data in the reviewed articles. The units of data in sediments and water were standardized as ng g^{-1} and ng L^{-1}, respectively.

3. Polychlorinated biphenyls

3.1. General information

PCBs are a group of aromatic chemicals that consist of a biphenyl structure bound to hydrogen and chlorine atoms. According to the number of hydrogen atoms substituted by chlorine atoms and their position on the biphenyl structure, the International Union of Pure and Applied Chemistry (IUPAC) has identified 209 congeners of PCBs (ATSDR, 2000). Being first synthesized by Oskar Gustav Doebner, a German chemist in 1876 and commercialized since the 1920s (Ngoubeyou et al., 2022), PCBs have been proven to

possess several crucial characteristics: having dielectric and electrical insulating properties, being resistant to chemical and thermal decomposition and non-flammable. Therefore, they had been used in electronics and hydraulic equipment, as plasticizers in paints, pigments, dyes, and rubber products (De & Pereira, 2004; Tue et al., 2013; Zhu et al., 2021). Due to their outstanding properties that were well served for industrial purposes, it was reported that from 1 to 1.5 million tons of PCBs had been produced globally since the 1920s, resulting in 17 million tons of products containing this group of chemicals (Othman et al., 2022). Little information could be found on the production of PCBs in Southeast Asia. PCBs were never produced in Indonesia, Laos or Vietnam (Falahudin et al., 2023; Kishida et al., 2007; Sudaryanto et al., 2011). In other Southeast Asian countries, while domestic production of PCBs has never reported, the widespread importation of PCB-containing equipment has significantly contributed to the regional contamination. Although most countries have implemented import ban on PCBs during the late 1990s to early 2000s, it was not until in 2016 did Indonesia last record the import of waste oil and electronics containing PCBs to the country (UNEP, 2021b). As of 2017, an inventory survey in Vietnam reported the presence of over 1300 tons of PCBs-contaminated oil; along with 26537 discarded capacitors and 35948 other pieces of equipment suspected to contain PCBs (UNEP, 2017). Overall, it is estimated that hundreds and thousands of tons of PCB-related materials remain across Southeast Asia, either in storage, still in use, or awaiting disposal.

Because of their wide applications, PCBs have been released into the environment through anthropogenic activities and detected in water, soil, sediment, air, aquatic species and human fluids due to their high resistance to degradation, bio-accumulative and persistent nature (H. Q. Anh et al., 2020; Ikemoto et al., 2008; Minh et al., 2004; Ngoubeyou et al., 2022; Thao, Kawano, et al., 1993). Early studies in the region reported the concentration of PCBs as equivalents to the commercial industrial mixtures of PCBs such as Aroclor 1254, Aroclor 1260 and Kanechlor (Duch Nhan et al., 1998; Viet et al., 2000). Later research used the sum of quantified PCBs congeners, especially 7 indicators PCBs (PCB 28, 52, 101, 118, 138, 153, and 180) suggested by the International Council for the Exploration of the Sea (7 ICES congeners, *Annex 2*) and 12 dioxin-like PCBs (PCB 77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169 and 189) suggested by World Health Organization (WHO).

This chlorinated group of chemicals exhibits many toxicological risks to both organisms and human health when accumulating in the trophic levels such as acting as carcinogens, disrupting reproductive function, causing dermatological adverse effects and developmental deficits, affecting the endocrine systems and hormonal levels (ATSDR, 2014). Given those potential effects on human bodies, the Stockholm Convention on Persistent Organic Pollutants listed, in 2004, PCBs as one of the twelve initial POPs and put them in Annex A (Elimination) suggesting that all member parties ban the production and new uses of PCBs. Furthermore, the Convention expects its members to implement waste management of all liquid waste and PCBs-contaminated products no later than 2028. Despite the efforts of governments, the United Nations Environment Program (UNEP) estimated, in January 2016, that only 17% (equivalent to 3 million tons) of PCB-containing materials and products have been eliminated resulting in more than 80% still in the environment on the global scale (UNEP, 2016a). Even though PCBs, as well as organochlorinated pesticides, had been phased out for years and regulated since the start of the Stockholm Convention, traces of them are still found in fresh and coastal waters in European countries (Castro-Jiménez et al., 2008; Kanzari et al., 2012; Liber et al., 2019), even though these countries are ranked as having the capacity to apply the convention in full. This implies that PCBs continue to leach into the environment and pose ecological and epidemiological threats.

3.2. Occurrence, concentrations and distribution in the abiotic aquatic compartment

While scientific research on the presence of PCBs in the region dates to the early 1990s, it was not until 2004 when the Stockholm convention included this group of chlorinated chemicals on the list of industrial POPs, and in the late 2000s, when the Southeast Asian countries signed the Convention, that comprehensive monitoring of PCBs in the Asian environment gained more attention (*Figure 13*).

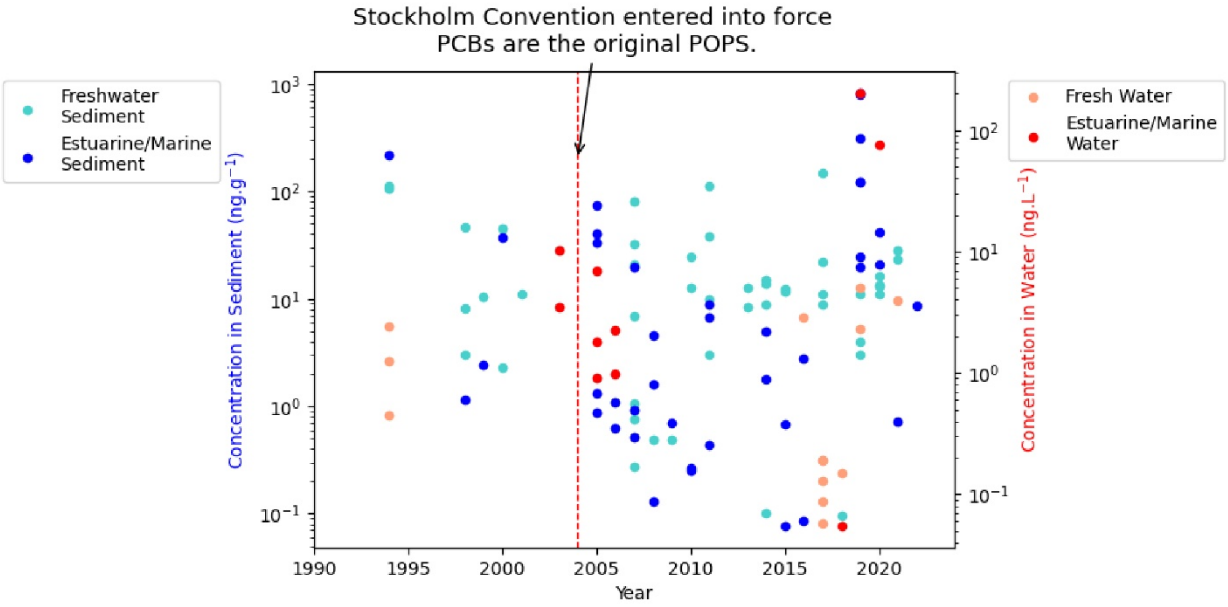

Figure 13: *Average PCBs concentration in freshwater sediment (n=46), estuarine/marine sediment (n=40), fresh water (n=13), and estuarine/marine water (n=10) over the studied period (1990-2024)*

For these monitoring programs, sediment samples from freshwater, estuarine and marine water bodies received more attention than water samples. Regardless of the ban from Southeast Asian countries since the early 2000s and the efforts of the Convention to identify and manage the PCB-containing products by 2028, the concentration ranges in environmental samples remained quite elevated and showed no sign of decline throughout the studied period (*Figure 13*). This trend may imply the continuous release of PCBs to different environmental matrices, especially from the storage sites for PCBs-containing fluids and equipment.

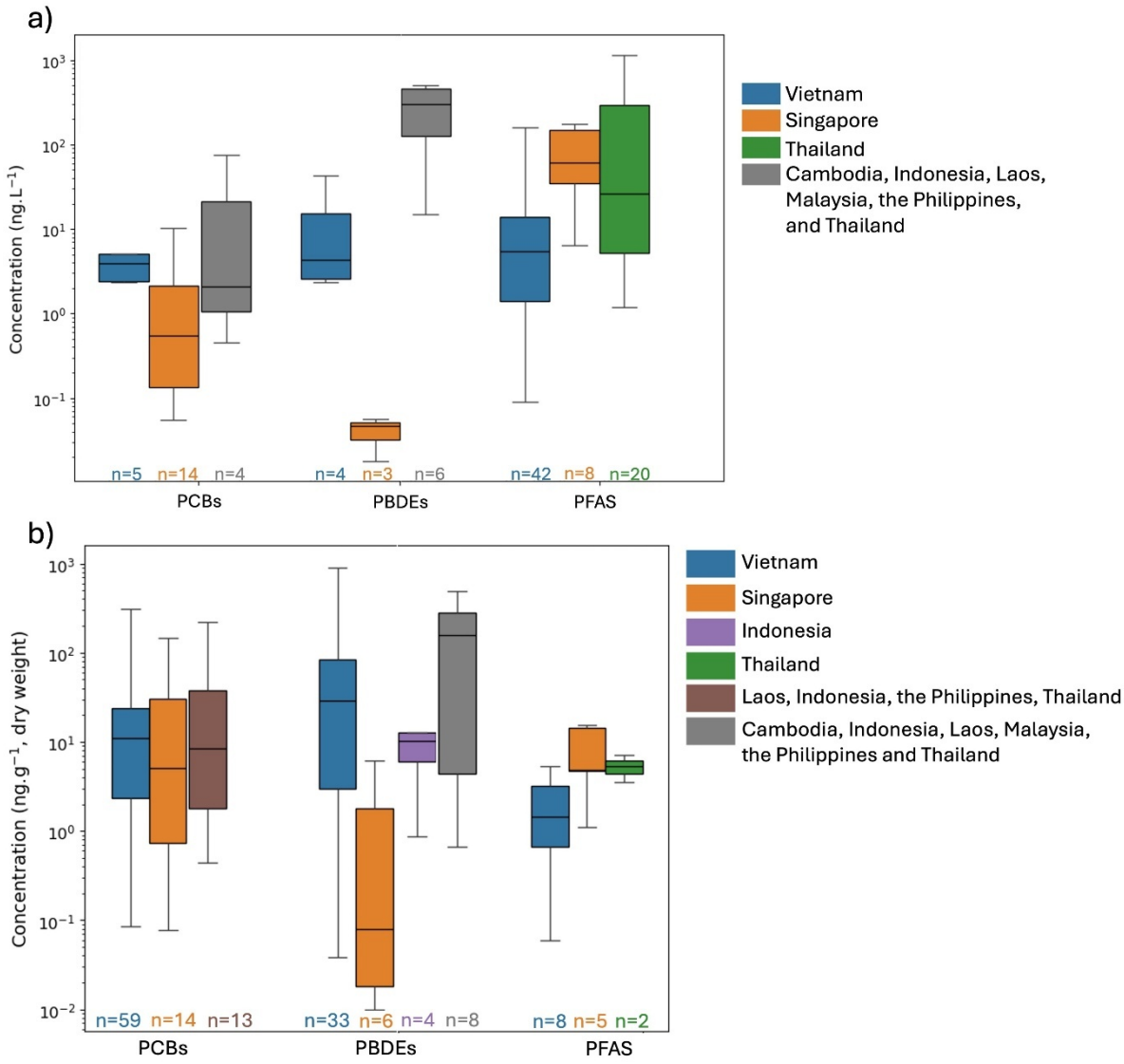

Figure 14: Reported average PCBs, PBDEs, and PFAS concentration in a) water and b) sediment samples in Southeast Asian countries.

Figure 14 shows that in the Southeast Asian region, the occurrence of PCBs in environmental aquatic compartments, especially in sediment, was mostly investigated in Vietnam, followed by Singapore and then other countries like Laos, Indonesia, the Philippines, and Thailand. For water samples (*Figure 14a*), the PCB levels reported in Singapore were lower when compared to those observed in other countries in the region. In contrast, the PCB concentrations in sediment (*Figure 14b*) appeared to be relatively consistent across the investigated nations. Under the time frame of this review, the earliest scientific research was conducted on water samples from the Klang River in Malaysia (Tan et al., 1991). The average concentration of PCBs in seven sampling stations

reached up to 1970 ng L^{-1}. Iwata et al (1994) carried out extensive research to investigate the occurrence of PCBs in water and sediment samples collected from sewage systems, canals, lakes, and rivers located in agricultural, urban, and industrial areas in Vietnam, Thailand Indonesia, and Malaysia. The total concentration of PCB isomers and congeners in sediment samples from Vietnam (n=18), Thailand (n=4) and Indonesia (n=4) could reach up to 630, 520 and 220 ng g^{-1} dw, respectively. The concentrations in water had a similar pattern to those of sediment with an average value of 2.4 ng L^{-1} for Vietnam (n=7), 1.77 for Thailand (n=5), 1.26 for Indonesia (n=3) and 0.45 for Malaysia (n=1).

Metropolitan areas constantly suffer from anthropogenic pollutions. The concentration of PCBs in the urban canals in Vientiane, the capital city of Laos (840,000 inhabitants in 2023) was in the range of 11 – 300 ng g^{-1} dw (Sudaryanto et al., 2011). A series of research focused on the densely populated areas of Hanoi, the capital city of Vietnam (4 million urban inhabitants in 2021). A large network of canals and urban rivers, including the Nhue River, To Lich River, Lu River, Set River, and Kim Nguu River received wastewater mainly coming from residential areas in the city. The contamination of PCBs in sediments from these rivers has been documented since the early 2000s with the concentration ranging from 0.3 to 96 ng g^{-1} dw (Duc Nhan et al., 2001; Hoai et al., 2010; Hoang, Takahashi, Da Le, Duong, et al., 2021). Furthermore, Kim Nguu River was investigated for the seasonal impact on the occurrence of PCBs in water and sediment samples (Quynh & Toan, 2019; Toan et al., 2020). It was pointed out that PCBs' concentrations were lower in the rainy season than in the dry season, implying that the rising rainfall could explain the dilution of contaminant burden in both water and sediment. Similarly, Minh et al. in 2007 reported the contamination of PCBs in sediment samples collected in urban canals and rivers in Ho Chi Minh City, another major city in Vietnam (7.2 million urban inhabitants in 2021). PCBs' concentrations were more elevated upstream with densely populated areas (average concentration of 81 ng g-1,dw in city canals and 6.9 ng g^{-1} dw in upstream river) and tended to decrease downstream, towards the ocean (average concentration of 0.9 ng g^{-1} dw in estuary zone). This conclusion on the spatial distribution in Sai Gon – Dong Nai River was in agreement with the Mekong River (Sudaryanto et al., 2011) and Kali Mas River (Ilyas et al., 2013). Nonetheless, 16 sediment samples and 183 water samples collected in an urbanized watershed in Singapore (5.9 million urban inhabitants in 2023) were reported with

relatively lower concentrations of PCBs when compared to big cities in Vietnam (Hanoi and Ho Chi Minh City) or Laos (Vientiane), with an average value of 23.81 ng g^{-1} dw and 0.15 ng L^{-1} respectively (Q. Wang & Kelly, 2017).

Compared to the highly populated zones above, marine waters appear to be less contaminated. PCBs were detected in several Singaporean marine waters but at lower concentrations than in urban waters, from less than 0.1 to just over 60 ng L^{-1} (Basheer et al., 2003; Bayen et al., 2003; Wurl & Obbard, 2005, 2006). Despite its proximity to metropolitan Manila, home to over 14 million inhabitants in 2023, the average concentration of PCBs found in 14 sediment samples from Manila Bay was less than 1 ng g^{-1} dw (Carvalho, Villeneuve, Cattini, Tolosa, et al., 2009; Villeneuve et al., 2010). Moreover, Yudhistira Azis et al. (2021) detected only PCB 28, 51 and 101 with an average concentration of 0.72 ng g^{-1} dw in seven sediment samples collected in the coastal mangrove forest of Indonesia. Even lower concentrations (36 - 158 pg g^{-1}, dw) were reported in surface sediments sampled from the offshore waters in the central region of Vietnam (Manh Tri et al., 2016). However, the case was not the same for Vietnam's coastal areas. When measuring PCBs in water from Cua Dai estuary, central Vietnam, Nguyen et al. (2019) detected high concentrations of 7 indicator PCBs with a maximum value of 1050 ng L^{-1}. PCBs were also found at high concentrations in sediment collected from the estuaries of Cua Hoi, Nhat Le and Lang Co Lagoon (the average values were 820, 799 and 121 ng g^{-1}, dw, respectively) in the same region (Tham et al., 2019).

The PCBs concentrations in freshwater samples reported in Southeast Asia satisfied the water quality guideline (14 ng L^{-1}) proposed by the United States Environmental Protection Agency (US-EPA, 2024b). But the levels of PCBs in marine waters were higher than the US-EPA standard for seawater, which is set at 3 ng L^{-1}. Some countries in the region also have certain regulations regarding PCBs in water. DENR Administrative Order 2016-08 of the Philippines government defined the classification of fresh water supplies and set the limit of PCBs in water for public use to be less than 100 ng L^{-1}. The concentration of PCBs has to be less than 500 ng L^{-1} in drinking water in Cambodia and in surface water in Vietnam (regulations listed in *Annex 5*).

3.3. Occurrence and concentrations in the aquatic trophic chain

Mollusks, i.e., filter feeders, are highly capable of accumulating contaminants in their tissues. Bayen et al. (2007) designed an experiment in the marine area of Singapore investigating the accumulation rate of oysters in an environment contaminated with PCBs, PBDEs, and DDT and chlordane pesticides in 2007. The oysters were cultivated for 230 days, and the oyster samples were analyzed for PCBs on the 0th, 126th and 230th day. Oysters raised in the uncontaminated sites witnessed better growth and higher tissue yield and lower levels of organic contaminants than those in contaminated sites. This finding signaled threats to the cultivation of oysters and other species in general, and it also represented the threat to humans when consuming contaminated aquacultural products.

The Asian Mussel Watch Program revealed an elevated level of PCBs in mussels collected in the urban and industrialized zones, reaching up to 1400 and 1900 ng g^{-1} lipid weight in samples from Indonesia and the Philippines, respectively (Ramu et al., 2007).

Additionally, signs of bioaccumulation and biomagnification were observed according to the detection of PCBs organisms of higher trophic levels than the filter feeders (*Table 4*), for example fish (Duc et al., 2024; Q. Wang & Kelly, 2018); green turtle eggs (van de Merwe et al., 2009), and whale and other cetaceans (Falahudin et al., 2023; Kajiwara et al., 2006). Monirith et al. reported the PCBs daily intake of Cambodians from fish to be 14 ng $person^{-1}$ day^{-1} in 1999, which was significantly lower than the acceptable daily intake set Food and Agriculture Organization (FAO) and WHO in 1986 (60,000 ng $person^{-1}$ day^{-1}). In 2003, WHO set a new tolerable daily intake of PBCs at 20 ng kg^{-1} body weight day^{-1} (WHO, 2003). Villeneuve et al. (2010) estimated the intake of 8.5 ng PCBs per kg body weight per day through the consumption of oysters by the Manila population in the Philippines. The consumption of marine fish in Vietnam would result in the intake of 3.04 to 6.92 ng PCBs per kg body weight per day, as calculated by Tran-Lam et al. (2023). The levels of PCBs in organisms from lower trophic levels (mussels, oysters, fish) do not pose significant health risks, as the estimated intake through their consumption remains below the acceptable daily intake thresholds (Monirith et al., 2000; Tran-Lam et al., 2023; Villeneuve et al., 2010). However, van de Merwe et al. (2009) warned that the consumption of a single turtle egg could result in an intake of coplanar PCBs (0.6 ng kg

body weight^{-1} day^{-1}) up to 300 times higher than the acceptable daily intake (ADI) established by the FAO/WHO in 2007 for coplanar PCBs (0.002 ng kg body weight^{-1} day^{-1}). This highlights a significant potential health risk associated with the consumption of contaminated products from wild animals, which is believed to have medicinal qualities by the local people in Malaysia. This would imply the growing concerning health risk with higher trophic levels, as biomagnification can lead to increased concentrations of PCBs in top predators exposed to human through consumption.

Table 4: Concentrations of PCBs (ng g^{-1}) measured in aquatic biota samples of the Southeast Asian region tabulated from lower to higher trophic levels.

Samples	Countries	Average concentration	Analytes	References
Plankton (*Planktothricoi-des spp and Synechococcus)*	Urban catchment in Singapore	826.5 ng g^{-1} lipid weight	28 PCBs	(Q. Wang & Kelly, 2018)
Apple snail (*Ampullariidae)*		420.6 ng g^{-1} lipid weight		
Clam (*Meretrix meretrix*)	Coastal zone, Northern Vietnam	7.88 ng g^{-1} dry weight	6 PCBs	(Duc Nhan et al., 1999)
Green mussels (*Perna viridis*) and blue mussels (*Mytilus edulis*)	Vietnam	76.32 ng g^{-1} lipid weight	62 PCBs	(Ramu et al., 2007)
	Cambodia	250 ng g^{-1} lipid weight		
	Indonesia	792.5 ng g^{-1} lipid weight		
	Malaysia	88.8 ng g^{-1} lipid weight		
	The Philippines	1500 ng g^{-1} lipid weight		

Oysters (*Crassostrea gigas*)	Manila Bay, the Philippines	19.89 ng g^{-1} dry weight	13 PCBs	(Carvalho, Villeneuve, Cattini, Tolosa, et al., 2009)
Clams	Mekong River	6.98 ng g^{-1} dry weight	13 PCBs	(Carvalho, Villeneuve, Cattini, Thuan, et al., 2009)
Skipjack Tuna (*Katsuwonus pelamis*)	The Philippines	190 ng g^{-1} lipid weight	117 PCBs	(Ueno et al., 2003)
	Indonesia	50 ng g^{-1} lipid weight		
Peacock bass *(Cichla orinocensis)*	Urban catchment in Singapore	3309 ng g^{-1} lipid weight	28 PCBs	(Q. Wang & Kelly, 2018)
Tilapia (*Oreochromis mossambicus)*		1238.2 ng g^{-1} lipid weight		
6 species of fish	Lagoon, Central Vietnam	34 ng g^{-1} lipid weight	7 PCBs	(T. A. M. Tran et al., 2019)
Various species of marine fish	Central-northern Vietnam	124 ng g^{-1} lipid weight	28 PCBs	(Duc et al., 2024)
Green turtle *(Chelonia mydas)* eggs	Malaysia	0.47 ng g^{-1} lipid weight	83 PCBs	(van de Merwe et al., 2009)
Cetaceans	The Philippines	3600 ng g^{-1} lipid weight	62 PCBs	(Kajiwara et al., 2006)
Short-finned pilot whale	Indonesia	237.9 ng g^{-1} lipid weight	209 PCBs	(Falahudin et al., 2023)

3.4. General profile of PCBs in environmental samples

The seven indicator PCBs (PCB 28, 52, 101, 118, 138, 158, and 180) have been widely detected in water, sediment and biota samples in the region. These congeners are often chosen as target analytes due to their presence in the technical mixture used in various industrial applications. Correlation tests and cluster analysis were performed to determine the relation between the congeners detected in samples and the components of the historically used commercial mixtures. Hoang et al. (2021) indicated that the profile of PCBs in sediment samples highly related to those in the commercial mixtures such as Sovol, Aroclor 1254, and Kanechlor 500. The pattern of Aroclor PCBs (PCB 101, 118, 153, and 138) was also found to be predominant in aquatic organisms in highly urbanized catchment in Singapore (Q. Wang & Kelly, 2018). As shown in *Figure 13*, despite the implementation of the Stockholm Convention and the ban on importation of PCBs in the Southeast Asian nations, the concentrations of PCBs in aquatic compartments show no decreasing trend. Given the quantity of PCBs inventoried in each country, the remaining stock of this chlorinated POPs remains significant and continues to be released into the environment.

4. Polybrominated Diphenyl Ethers

4.1. General information

Polybrominated Diphenyl Ethers (PBDEs) are a group of organic chemicals consisting of a diphenyl ether structure bound to bromine atoms. Similarly to PCBs, depending on the numbers of bromine atoms and their positions on the aromatic rings, IUPAC has identified 209 congeners categorized into 10 groups from mono- to decabromodiphenyl ethers (ATSDR, 2017). There had been about 1.5 million tons of PBDEs produced globally since the 1970s (Motamedi et al., 2022). Being highly resistant to physical and chemical degradation, PBDEs had been widely used as flame retardants in various products such as electronics, furniture, and textiles (Costa & Giordano, 2014; Tue et al., 2013). Three major commercialized PBDE mixtures are pentaBDE (composed of BDE 47, 99 and 100), octaBDE (BDE-183 and other octa-congeners) and decaBDE (mostly composed of BDE-209) (Kefeni et al., 2011). By 2005, the historical productions of penta-, octa-, and decaBDE mixtures were approximately estimated to be 0.175, 0.13, and 1.1 million tons, respectively (Abbasi et al., 2019; T. Meng et al., 2021). Similarly to PCBs, inventory efforts

on PBDEs have been carried out the Southeast Asian countries and reported under the framework of the Stockholm Convention. While no domestic production was recorded in these countries, the importation of electronics and electrical equipment (EEE) has emerged as a significant source of PBDE contamination. Laos reported in 2016 a stockpile of approximately 5.8 tons of commercial octa-BDE associated with imported household EEE (UNEP, 2016b). In Thailand, an estimated 920 to 1500 tons commercial deca-BDE were identified in 2022 in imported television sets, along with an additional 3 tons of deca-BDE found in flame-retardants fabrics (UNEP, 2022b). In addition to EEE, imported vehicles have also been identified as potential sources for PBDE contamination. In 2014, the Philippines recorded 2.2 tons of commercial penta-BDE on imported vehicles as well as additional 2 tons found in cars, trucks, and buses that are classified as end-of-life vehicles (UNEP, 2014). In parallel, in 2017, Vietnam reported more than 6 tons of PBDEs found in polymer components of vehicles that were still in use (UNEP, 2017).

These flame retardants are additive compounds, or in other words, they are only physically bonded to the products. Consequently, they can easily escape and leach into the environment (Costa & Giordano, 2014; Darnerud et al., 2001). Similarly to PCBs, PBDEs have been shown to exhibit severe toxicity towards humans; being endocrine disruptors and carcinogens, causing DNA damage and inducing neurotoxicity (Couderc et al., 2016; Motamedi et al., 2022; Renzelli et al., 2023). Because of the potential health impact on humans, the US and Europe banned the production of these brominated chemicals in the 2000s (Abbasi et al., 2019). According to Directive 2013/39/EU under the European Water Framework Directive (WFD), the maximum allowable concentration for six PBDEs (BDE 28, 47, 99, 100, 153, and 154) was 140 ng L^{-1} for inland surface waters (lakes, rivers and other artificial and heavily modified water bodies). In addition, the Stockholm Convention listed penta- and octaBDE mixtures in Annex A (Elimination) in 2009. And 8 years later in 2017, the Convention also added the decaBDE mixture to Annex A, banning its production and use, with specific exemptions for certain applications (e.g. textile productions requiring anti-flammable characteristics, additives in plastic housings and parts at concentrations lower than 10% by weight of the part, and certain vehicles and aircrafts, Decision UNEP/POPS/COP.8/10, https://chm.pops.int/Convention/COP/COPDecisions/tabid/208/Default.aspx, accessed March 2025). Besides the allowable limit of PBDEs content in electrical and electric products (0.1% by weight) set by Singaporean and Vietnamese governments, no other

regulations on PBDEs-containing products were documented in other Southeast Asian nations (Hoang et al., 2021).

4.2. Occurrence, concentrations and distribution in the abiotic aquatic compartment

PBDEs have received more attention after they were identified as POPs by the Stockholm Convention in 2009 for tetra- and pentaBDE and in 2017 for decaBDE. Throughout the years, PBDEs have been progressively searched for and detected in environmental samples with a wide concentration range in sediment and water (*Figure 15*). Quite similarly to PCBs, PBDE levels in sediment remain highly variable and show no clear signs of decline, despite existing regulatory measures. This persistent presence suggests that regulatory actions may not have made considerable impact, possibly due to the continued release from existing sources and the nature of high resistance to degradation of these compounds. For water, the PBDE levels appeared to be decreasing toward the late 2010s and early 2020s. However, the limited data on the levels of PBDE in water samples prevents making a definitive conclusion on the overall temporal trend.

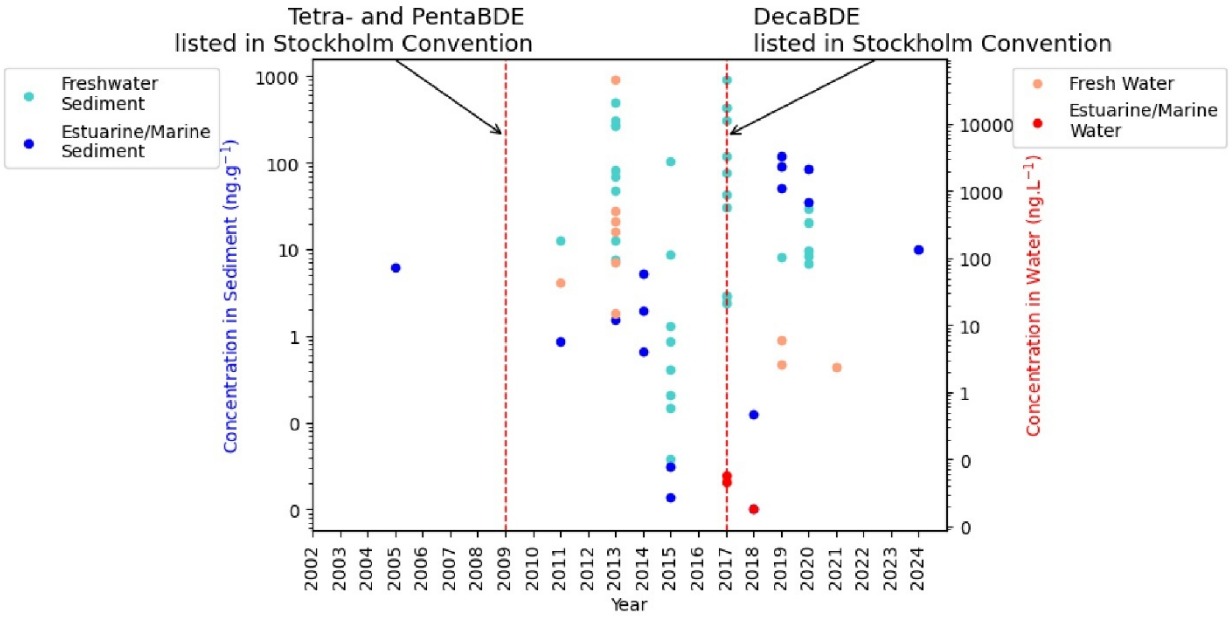

Figure 15: Average PBDEs concentration in freshwater sediment (n=35), estuarine/marine sediment (n=16), fresh water (n=10), and estuarine/marine water (n=3) over the studied period (1990-2024).

Generally, the reported data on studies in this region was the sum of different PBDEs congeners detected in environmental samples. As the three major mixtures consist mostly of BDE-28, -47, -99, -100, -153, -154, -183 and -209 (Salvadó et al., 2012), these eight representative congeners (*Annex 3*) are frequently measured for environmental monitoring.

Being hydrophobic with low water solubility and high n-octanol-water partition coefficient (4.3-9.9), PBDEs preferentially partition into particulate matter and gradually accumulate in sediment (Darnerud et al., 2001; Kwan et al., 2013). Therefore, it might explain why the occurrence of PBDE is better documented in sediment than in water for Southeast Asian countries (*Figure 14*). High urbanization and industrialization rates might lead to higher production of wastes, especially electronic waste, or e-waste, which contribute significantly to the presence of PBDEs in the environment. It was estimated that in 2019, Southeast Asian countries had generated three and a half megaton e-wastes (Forti et al., 2020). Although recycling e-wastes is considered a sound strategy for disposal and may bring economic benefits (Matsukami et al., 2015, 2022), the recycling sites where e-wastes are gathered could become significant sources of PBDEs released into the environment. Sediments collected from drainage canals in Trieu Khuc e-waste recycling village (Vietnam) were found to be contaminated with the eight PBDEs (BDE 28, 47, 99, 100, 153, 154, 183 and 209), whose average sum concentration reached up to 434 ng g^{-1} dw (H. Q. Anh et al., 2017). The authors also reported a contamination by the eight congeners not only in sediment but also in fish found in nearby ponds and canals of Bui Dau recycling village (Vietnam). More alarmingly, Matsukami et al (2005), who worked on the samples from the Bui Dau e-waste recycling village, recorded an increase in the concentration of PBDEs in sediment throughout a 3-year period. This highlights that even though PBDEs are no longer produced, the risk of environmental contamination remains high unless efficient treatment processes of e-waste are developed and followed to reduce the accidental release of PBDEs into the environment.

The screening of PBDEs in water was less common in the studied countries. Kwan et al. in 2013 conducted a screening of 46 PBDEs in leachates from municipal waste dumping sites in seven countries in the region. The leachates from dumping sites in the industrialized city of Bangkok (Thailand) showed extremely high concentrations, reaching up to 133,000 ng L^{-1}. The values were one to three orders of magnitude lower

for other less industrialized countries such as Laos, Cambodia and the Philippines. On the contrary, Wang & Kelly (2017) reported no further inputs of PBDEs to the environment, as they detected very low concentrations in water samples (average values of 0.046 and 0.057 ng L^{-1} in open water bodies (n=32) and in catchment channels (n=24), respectively) collected in an urban watershed in Singapore, which stands as the only developed country in Southeast Asia. This finding was consistent with the clear trend shown in *14* where PBDE levels in both water and sediment samples from Singapore were significantly lower than those reported in other developing Southeast Asian countries.

4.3. Occurrence and concentrations in the aquatic trophic chain

Having a lipophilic nature, PBDEs can gradually accumulate into living organisms (Darnerud et al., 2001; Falahudin et al., 2022). The earliest studies on the occurrence of PBDEs investigated, in the early 2000s, their presence in mussels (8.85 ng g^{-1} dw, Bayen et al., 2003) in skipjack tunas (14 ng g^{-1} lipid weight, Ueno et al., 2004) and in freshwater fish captured near dumping sites in the Mekong river delta (36 ng g^{-1}, lipid weight, Hung Minh et al., 2006). Later studies alerted on the risk of bioaccumulation and biomagnification of PBDEs in aquatic organisms from low trophic levels (plankton, barnacles, mussels) to higher ones (turtles, dolphins) (*Table 5).* The marine traffic and the industrial activities in the coastal areas could be responsible for the contamination of PBDEs in marine organisms (Ramu et al., 2007). US-EPA has derived the oral reference doses (RfD) for PBDEs as follows: 7000 ng body $weight^{-1}$ day^{-1} for decaBDE (BDE 209), 3000 ng body $weight^{-1}$ day^{-1} for octaBDE, 2000 ng body $weight^{-1}$ day^{-1} for pentaBDE, 100 ng body $weight^{-1}$ day^{-1} for both BDE 47 and 99, and 200 ng body $weight^{-1}$ day^{-1} for BDE 153 (ATSDR, 2017). People living in coastal areas in northern Vietnam could up take from 9.06 to 33.5 ng of 8 PBDEs body $weight^{-1}$ day^{-1} via fish consumption (BDE 28, 47, 99, 100, 153, 154, 183, and 209, Duc et al., 2024). Furthermore, elevated levels of PBDEs found in fish caught in the waterbodies near e-waste recycling sites in Vietnam (H. Q. Anh et al., 2017), once again, emphasized the significant source of contaminants due to improper handling of PBDEs-containing e-waste and called for the attention of authorities on the recycling and disposal activities. The daily intake of eight PBDE congeners (BDE 28, 47, 99, 100, 153, 154, 183, and 209) among residents of e-waste recycling sites was 154 ng $person^{-1}$ day^{-1} for children and 730 ng $person^{-1}$ day^{-1} for adults. BDE 209 contributed to more than 80% of total daily PBDEs intake in both groups.

Table 5: Concentrations of PBDEs (ng g^{-1}) measured in aquatic biota samples of the Southeast Asian region tabulated from lower to higher trophic levels.

Sample	Countries	Average concentration	Analytes	References
Green mussels (*Perna viridis*)	Singapore	8.85 ng g^{-1} dry weight	7 PBDEs	(Bayen et al., 2003)
Mussels (*Perna viridis*, *Mytilus edulis*)	Cambodia	3.81 ng g^{-1} lipid weight	9 PBDEs	(Ramu et al., 2007)
	Indonesia	13.2 ng g^{-1} lipid weight		
	Malaysia	6.7 ng g^{-1} lipid weight		
	The Philippines	103.8 ng g^{-1} lipid weight		
	Vietnam	2 ng g^{-1} lipid weight		
Barnacles		51.2 ng g^{-1} lipid weight	44 PBDEs	(Vaezzadeh, Thomes, et al., 2021)
Green mussels		60.5 ng g^{-1} lipid weight		
Green mussels (*Perna viridis*)	Vietnam	8.63 ng g^{-1} dry weight	5 PBDEs	(D. T. Nguyen et al., 2024)
Catfish (*Pangasianodon hypophthalmus*)	Mekong river delta, Vietnam	2.7 ng g^{-1} lipid weight	6 PBDEs	(Hung Minh et al., 2006)
Three species of carps	Vietnam	160 – 1380 ng g^{-1} lipid weight	8 PBDEs	(H. Q. Anh et al., 2017)
Various freshwater species	Urban catching in Singapore	48.4 – 474.5 ng g^{-1} lipid weight	7 PBDEs	(Q. Wang & Kelly, 2018)

Marine fish	Vietnam	67.7 ng g^{-1} lipid weight	8 PBDEs	(Duc et al., 2024)
Skipjack Tuna (*Katsuwonus pelamis*)	Philippines	14 ng g^{-1} lipid weight	11 PBDEs	(Ueno et al., 2004)
Green turtle (*Chelonia mydas)* eggs	Malaysia	21.4 pg g^{-1} wet weight	19 PBDEs	(van de Merwe et al., 2009)
Spinner dolphins (*Stenella longirostris*)	Philippines	36 ng g^{-1} lipid weight	10 PBDE	(Kajiwara et al., 2006)

4.4. General profile of PBDEs in environmental samples

Overall, the PBDE profiles in samples from aquatic compartment in Southeast Asia saw a predominance of congeners that are the most prevalent in the technical mixtures (i.e., BDE 47, 99, 100, 153, 154, 183, and especially BDE 209). BDE 209 is the main component of the commercial deca-BDE mixture, which accounted for nearly 80% of the historical production of PBDE mixture. Consequently, the widespread presence of imported products contaminated with PBDEs has significantly contributed to the environmental burden of these brominated POPs in the region. Furthermore, the detection of nona-BDE congeners (e.g., BDE 206, 207, and 208) in sediment, leachate, and biota samples has prompted further discussion on the debromination of PBDEs (Ilyas et al., 2011; Kwan et al., 2013). Higher-brominated PBDEs, particularly BDE 209, could be transformed under anaerobic conditions or through metabolic processes in organisms, leading to the formation of lower-brominated PBDEs (Ilyas et al., 2013; Q. Wang & Kelly, 2017). This debromination process may explain the presence of certain nona-BDE congeners in environmental samples, even though these are not commonly found in technical BDE mixtures.

5. Poly- and Perfluoroalkyl substances

5.1. General information

PFAS are a group of synthetic chemicals including carbon atoms and fluorine atoms covalently bonded to the alkyl chain (Buck et al., 2011) and different functional groups. It is estimated that over 8000 chemicals of known structures have been identified as PFAS (Evich et al., 2022). Because PFAS are highly resistant to extreme heat and repellent to oil and water, they are used in a variety of consumer products, for example in cookware, food packaging, paint, textile, and aqueous firefighting foam (Buck et al., 2011; Phong Vo et al., 2020; Schmidt et al., 2019).

Several countries in Southeast Asia have conducted inventory work for perfluorooctane sulfonic acid (PFOS). Quite similarly to the chlorinated and brominated POPs, PFOS was not produced in Southeast Asia but instead introduced into the region through the importation of finished products. Laos, until 2016, imported 366 tons of PFOS used in synthetic materials whereas the Philippines reported over 500 tons of PFOS incorporated in imported articles (i.e. synthetic carpets, fire resistant textiles, chrome metal-plating pigments and food packaging) only in 5-year period from 2008 to 2012 (UNEP, 2016b, 2022b). The presence of PFOS in firefighting foams is significant. In 2019, Thailand was estimated to have between 3.7 and 5.5 tons of PFOS contained in 925000 liters of firefighting foams held in stock. The Vietnamese inventory report in 2017 suggested the importation between 10 and 15 tons of PFOS in firefighting foams during the period from 1998 to 2013. Annual PFOS imports for other applications were comparable in the same time period, ranging from 0.11 to 3.45 tons for textiles and upholstery, and from 0.2 to 4.8 tons for paper and paperboard products. (UNEP, 2017).

Because of their wide applications, PFAS have become ubiquitous in the environment.

Toxic effects of PFAS are mostly attributed to endocrine disruption mechanisms leading to health outcomes (Braun, 2017) including epigenetic changes and a number of autoimmune diseases (e.g., leukemia, breast cancer, diabetes, multiple sclerosis) and other health problems (e.g., early puberty, reduced fertility (S. Kim et al., 2021). PFAS also exhibits toxicity, e.g., immunotoxic, dyslipidemic, neurotoxic effects (M. Bonato et al., 2020; Fenton et al., 2021). Of note, PFAS have been found in blood samples of almost all human population (Fenton et al., 2021), and induce oxidative stress, autophagy as well as

apoptosis at cell level in human (Zeng et al., 2021). Bioaccumulation have been highlighted for many organisms, including both PFAAs and few precursors (Alfaro Garcia et al., 2022; Evich et al., 2022; Herzke et al., 2023; Jouanneau et al., 2020; Macorps et al., 2022). PFAS biomagnification has also been evidenced for longer chain Perfluoroalkyl acids (PFAAs), whereas several precursors were not found to be magnified along trophic levels, due to potential metabolization (Simonnet-Laprade et al., 2019). Cell toxicity and endocrine disruption has also been shown for aquatic species (Hu et al., 2003; Shi et al., 2019). Another issue is the apparent multigenerational effects of PFAAs exposure (Z. Li et al., 2020). Remarkably, most studies focus on PFAAs effects, and the literature on precursors effects is still scarce (Macorps et al., 2022; McDonough et al., 2022; Panieri et al., 2022). Moreover, PFAS occur as a mixture of PFAAs and precursors in the environment, and these mixture effects have even less been investigated (Dickman & Aga, 2022).

Perfluorooctanoic acid (PFOA) and PFOS are the most regulated PFAS. The Stockholm Convention has listed PFOS in 2009 in Annex B (Restriction) and PFOA, in 2019, in Annex A (Elimination). Moreover, in 2019, the Convention banned the production of PFOS, its salts and perfluorooctane sulfonyl fluoride; and set specific exemptions on their usage on metal plating and fire-fighting foam (Decision UNEP/POPS/COP.9/4, https://chm.pops.int/Convention/COP/COPDecisions/tabid/208/Default.aspx, accessed March 2025). Perfluorohexane sulfonic acid (PFHxS) and its salts were added into the same Annex as PFOA in 2022. Furthermore, the European Commission, in Directive 2013/39/EU, set the annual average environmental quality standard (AA-EQS) for PFOS and its derivatives at 0.65 and 0.13 ng L^{-1} for inland and other surface waters, respectively. Regulatory agencies worldwide have established restrictions for PFAS levels in drinking water. The Drinking Water Directive (DWD-EU) 2020/2184 of the European Parliament introduced a limit for the sum concentration of 20 PFAS (10 perfluoroalkyl carboxylic acids and 10 perfluoroalkyl sulfonic acids) at 100 ng L^{-1}. More recently in April 2024, the US-EPA announced the National Primary Drinking Water Regulation (NPDWR) for several PFAS, three of which are listed in the Stockholm Convention (PFOA, PFOS and PFHxS). The NPDWR indicates the limits for PFOA and PFOS each at 4 ng L^{-1} and for PFHxS at 10 ng L^{-1} in drinking water (US-EPA, 2024a).

5.2. An emerging concern in South-East Asia

Similarly to PCBs and PBDEs, the 2010s witnessed an increase in PFAS research articles conducted in Southeast Asia (*Annex 7*). While only one article on PFAS could be found before PFOS was listed in the Stockholm Convention, 18 new articles were found 10 years after the regulations on PFOS and 15 more were published during a four-year period after the listing of PFOA. The number of PFAS research in Southeast Asia is comparable to that of Japan (27 articles) and Korea (37 articles) in East Asia, but it is approximately 5 times lower than in China (163 articles) (Tang et al., 2023).

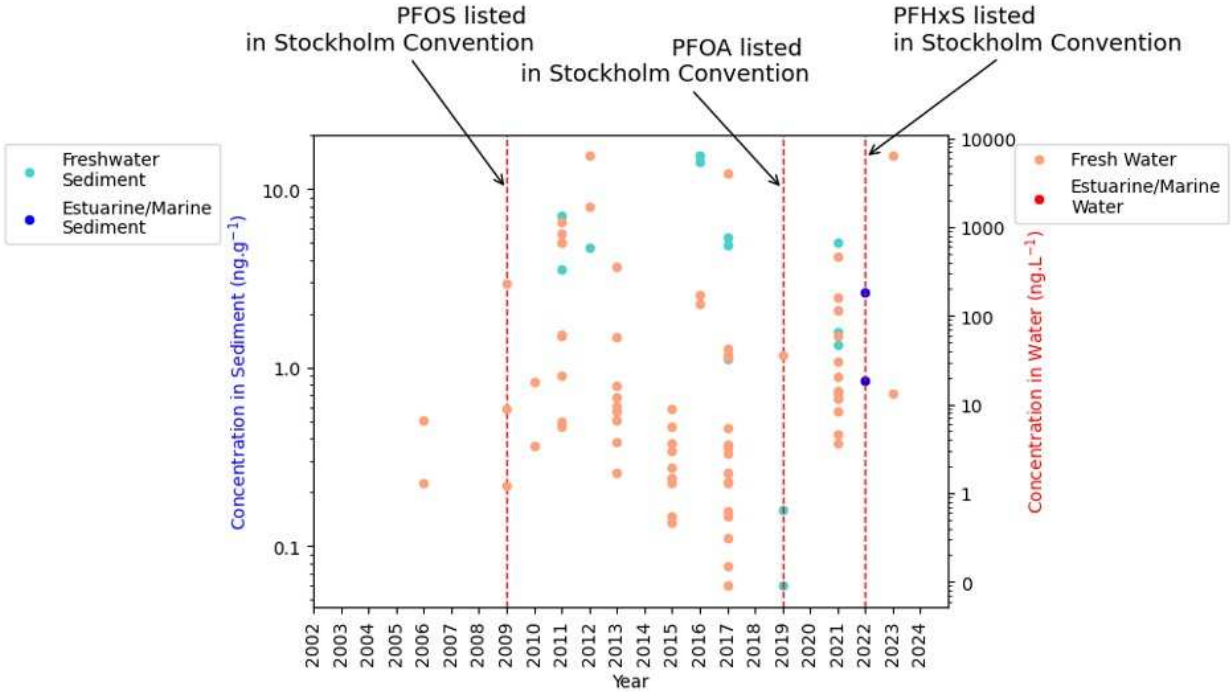

Figure 16: Average PFAS concentrations in freshwater sediment (n=13), estuarine/marine sediment (n=2), fresh water (n=70) over the studied period (1990-2024).

Figure 16 presents the data on PFAS in water and sediment samples in Southeast Asia. The observed concentrations span a wide range, reflecting the variability across sites and sample types. Despite the implementation of PFAS regulations, there is no evident decreasing trend in environmental concentrations. This may be explained by the fact that monitoring activities in the region only began after the introduction of these regulations. Besides the regulations established by the Stockholm Convention, the Southeast Asian nations have not yet introduced their own national limits for PFAS concentration in various matrices. Efforts to establish comprehensive inventory and identify the PFAS

occurrence are relatively recent, which may be limitations for the investigations of long-term trends.

5.3. Occurrence and concentrations in waters and edible aquatic species

The studies on PFAS in the environment were only conducted in the Philippines, Singapore, Thailand, Vietnam, and Malaysia and mostly focused on the water samples (*Table 6 and Figure 14*). The early 2010s marked the pioneering studies on PFAS in the region. The Langat River in Malaysia was reported with important levels of PFOA and PFOS (respective maximum concentrations at 5,940 and 43,500 ng L^{-1}) at sampling stations of a landfill area and a creational camping site. The results were coherent with the detection of PFOA and PFOS in the leachate samples collected in landfills in Hanoi (J. W. Kim et al., 2013; Ngoc et al., 2021) and in Singapore (Yin et al., 2017). Therefore, it is of importance to apply proper and efficient treatment methods for leachate, and wastewater in general, to prevent the dissemination of PFAS through water transport. However, not only might the conventional methods in wastewater treatment plants be ineffective in completely removing PFAS, but the total concentration of PFAS in effluents may also increase, likely due to the degradation of precursors forming new PFAS, particularly PFOA and PFOS (Kunacheva et al., 2010, 2011). On the contrary, Tabtong et al. (2015) concluded on the removal rate of 86% of PFAAs when advanced treatment methods using granular activated carbon filter and reverse osmosis are applied.

The presence of PFAS was recorded in sediment samples collected in urban water bodies in Singapore (T. V. Nguyen et al., 2016; V. T. Nguyen et al., 2012) and in large cities in Vietnam (Babut et al., 2019; Ngoc et al., 2021; H. Le Tran et al., 2022) with the average concentrations ranging from less than 1 up to 15 ng g^{-1}, dry weight (*Figure 14*).

More alarmingly, PFAS have been detected in drinking water, bottled water, and biota samples (*Table 6* and *7*). The presence of PFAS in bottled water in Thailand highlighted the inefficiency of the treatment methods (Kunacheva et al., 2010; Lertassavakorn et al., 2021). This emphasizes the potential exposure risk for humans to these organic contaminants. Other than contact with PFAS-containing products (for example, cookware and food packaging), the potential consumption of contaminated water or food indicates a direct exposure route via the digestive system, thus making it significantly dangerous for human health. The levels of PFAS in drinking water samples collected in the

Philippines, Thailand and Vietnam were well below European and American limits (*Table 6*). *Table 7* signals the accumulation of PFAS in aquatic organisms, especially species usually served as food. The European Food Safety Authority established the tolerable weekly intake (TWI) of four PFAS (PFOA, PFNA, PFHxS, and PFOS) to be 4.4 ng kg body weight^{-1} week^{-1} (Schrenk et al., 2020). Murakami et al. (2011) reported the daily intake of both PFOS and PFOA to be 0.51 ng kg body weight^{-1} day^{-1} for population in some Asian countries, including Malaysia and Vietnam. The maximum weekly intake of PFOA, PFNA, PFHxS, and PFOS for population in the Red River delta (Vietnam) was calculated to be 3.7 ng kg body weight^{-1} week^{-1}, which was still lower than the European limit (Hoa et al., 2022). On the other hand, study carried out by Lertassavakorn et al. (2021) showed that the intake of only PFOS was 1.9 ng kg body weight^{-1} week^{-1} for general population but could reach up to 37.42 ng kg body weight^{-1} week^{-1} for the consumers of seafood living close to a coastal industrial zone in Thailand. The authors even advised against the regular consumption of squid and spotted babylon to avoid the potential health risks related to PFAS exposure. Even if some of the reviewed studies on biota samples in Southeast Asia alarmed no immediate risks with fish consumption, as the daily intake lied under the acceptable thresholds, long-term exposure to PFAS, accumulating from both drinking water and seafood, would still pose significant health concerns.

Table 6: Concentrations of PFAS (ng L^{-1}) measured in tap, ground, drinking and bottled waters of the Southeast Asian region.

Country	Sample type	Concentration (ng L^{-1})	Analytes	Reference
Philippines	Drinking water	9.08 – 11.63	12 PFAS	(Guardian et al., 2020)
Thailand	Tap water	10*	8 PFAS	(Kunacheva et al., 2009)
	Tap water	6.03 *	5 PFAS detected	(Kunacheva et al., 2010)
	Bottled water	11.64 *		
	Tap water	0.4*	PFOS	(Boontanon et al., 2013)
	Bottled water	0.5*		
	Drinking water	7.16 – 59.49	12 PFAS	(Guardian et al., 2020)
	Tap water	54.2 **	PFOS	(Lertassavakorn et al., 2021)
	Bottled water	0.45	PFOS	
		0.62	PFOA	
	Groundwater	13.4 *	6 PFAS detected	(Hongkachok et al., 2023)
Vietnam	Groundwater	0.54-5.56	7 PFAS detected	(Duong et al., 2015)
	Tap water	0.31 *	13 PFAS	(Lam et al., 2017)
	Well water	3.55 *		
	Drinking water	0.42	PFOA	(Tröger et al., 2021)
		0.26	PFHpA	
		0.13	PFNA	

*: average values, **: maximum values

Table 7: Concentrations of PFAS (ng g^{-1}) measured in aquatic biota samples of the Southeast Asian region.

Samples	Countries	Concentration (ng g^{-1} wet weight)	Analytes	References
Snakehead (scientific name not identified)	Malay (n=2)	Liver: 4.6 * Muscle: 0.4 *	10 PFAS	(Murakami et al., 2011)
	Vietnam (n=4)	Liver: 23.18 * Muscle: 0.33 *		
Various aquatic species	Vietnam	0.33 - 2.81	13 PFAS	(Lam et al., 2017)
Four common food fish species (*Hypophthalmic-hthys nobilis*, *Cyprinus carpio*, *Labeo rohita*, and *Tilapia spp*.)	Vietnam	Blood: 5.2 – 29 ng mL^{-1} Liver: 2.7 – 6.6 Muscle: 0.51 – 2.6	17 PFAS	(Hoa et al., 2022)
Freshwater fish	Vietnam	0.08 – 8.06	9 PFAS	(Vi et al., 2022)
Marine fish		0.22 – 3.62		
Various marine species	Thailand	< LOD – 6.724	PFOS, PFOA	(Lertassavakorn et al., 2021)
Marine fish	Vietnam	9 – 28.6	23 PFAS	(Vu-Duc et al., 2023)

*: average values

5.4. PFAS and the analytical trends

PFOA and PFOS are the first two PFAS to be listed as POPs in the Stockholm Convention. During the 2000s and 2010s, PFAS research in Southeast Asia primarily focused on these two compounds, along with other PFAAs. The inventory work done in this time period included only PFOS, given its widespread use in textiles and firefighting foams across the region. Following the implementation of new regulations on the production and use of PFOA, PFOS, PFHxS, alternative PFAS compounds began to emerge as replacements for

the restricted substances in industrial processes. For example, 6:2 fluorotelomer sulfonic acid (6:2-FTS) is used as substitutes for PFOS in metal electroplating, and N-Methyl Perfluorooctane sulfonamidoethanol (N-MeFOSE) and N-Ethyl Perfluorooctane sulfonamidoethanol (N-EtFOSE) are used paper, packaging and textile products (Brunn et al., 2023; Göckener et al., 2021a). As a results, more recent researches have also taken these emerging PFAS into account. 6:2-FTS, N-Methyl Perfluorooactane sulfonamidoacetic acid (N-MeFOSAA), and N-Ethyl Perfluorooctane sulfonamidoacetic acid (N-EtFOSAA) have been included in the list of targeted analytes and detected in surface water, sediment and fish samples (H. Chen et al., 2019; Vu-Duc et al., 2023). H. Chen et al. even reported the dominance of EtFOSAA over legacy PFAS (i.e. PFOS, PFOA) in the surface sediment samples collected in 2017 in Singapore. Although the regulatory framework for PFAS management in Southeast Asia is still under development and has not yet been comprehensively established, the region's scientific community has demonstrated a proactive approach in responding to emerging PFAS challenges. Research groups have kept pace with evolving trends in PFAS usage, including the shift towards alternative compounds, and have been adapting monitoring strategies accordingly.

6. Research gaps and future directions

6.1. Economic growth and extreme climate events

Studies of PCBs and PBDEs have been carried out since the 1990s in eight Southeast Asian countries. They have received much more attention since 2004 and 2010, after the regulatory agencies, not only regional but also worldwide, take effective measures to mitigate their negative impacts. The levels of PCBs and PBDEs in the SEA environment were comparable to those in the Bohai Sea and Yellow Sea in China and South Korea (J. Meng et al., 2017; H. Yu et al., 2023), in the Mediterranean Sea in Southern Europe (Kanzari et al., 2012, 2014), and in North America environment (Hale et al., 2003). Although PCBs and PBDEs have been successively banned, their replacement has involved the marketing of products with similar potential applications. This gives currently rise to the production of very diverse brominated and chlorinated molecules, some of which have characteristics similar to POPs and merit specific attention from the scientific community (Rigal et al., 2024; Vénisseau et al., 2018). In comparison to PCBs and PBDEs, PFAS were poorly investigated in South-East Asia. Hence further research is required to have a more comprehensive understanding of PFAS contamination in this region. This is of high human

health concern, since PFAS have been detected in drinking waters and various aquatic species.

Moreover, it is utmost important to conduct regular monitoring of POPs to gain a better understanding on the change in quality status of the water resources, owing to two cumulative factors taking place in the region (H. Zhang et al., 2019). On the one's hand, the rapid economic growth is likely to intensify the use of chemicals and the consumption of consumer goods. The rapid economic and population growth act driving factors for the increased rate of production and consumption. The PBDEs emission is proven to be correlated with population growth, urbanization and industrialization (Kwan et al., 2014). A higher GDP per capita often reflects increased purchasing power and higher living standard, which can eventually lead to greater demand for goods and services. Majority of these products, such as electronics, vehicles, textiles, and household appliances, as indicated in the inventory reports, contain POPs. The region with fast economic development, therefore, may exhibit increased potential for POP emissions and accumulation in the environment. In addition, climate change will lead to more frequent extreme events, which should result in the remobilization of sedimentary pollutants and long-range transport to remote areas (Nadal et al., 2015; Pizzini et al., 2022; Teran et al., 2012). Santiago & Rivas (2012) reported higher PCBs levels in water after typhoon due to the overflow from river and runoff from surrounding areas. Similarly, Lertassavakorn et al., (2021) suggested that the heavy precipitation events could contribute to the repartition of PFOA and PFOS, leading to more elevated concentration in environmental samples. These conclusions were further supported by the results from climate models, which suggested that the increase in precipitation would lead to higher levels of POPs in environmental compartments (Noyes et al., 2025). Moreover, climate change affects not only abiotic environmental matrices but also has significant impacts on biotic components. Living organisms, including wildlife, are becoming more vulnerable to the change in climate patterns, which can eventually influence their exposure to and accumulation of organic contaminants. Foster et al. (2019) showed clear correlations between rainfall and concentrations of organochlorine pollutant in seabirds. The study pointed out that increased concentrations of PCBs in seabird eggs were associated with higher rainfall. This emphasizes once again the link between the intensified precipitation and the enhancement of mobilization and bioavailability of POPs. The findings of climate models highlight the complex interactions between the extreme events caused by climate

changes and the dynamics of POPs in environmental systems. In this context, the fate of POPs must be followed more systematically in Southeast Asia, given the region's vulnerability to climate change. The scientific findings would be helpful for policy makers to set national standards in water, sediment, and biota samples and to prevent potential ecological and human health risks. On the other hand, the development of proper treatment protocols for the discharge or recycling of POPs-containing products, like e-product or plastic, is needed to limit POP uncontrolled dispersion into the aquatic resources and to achieve the goals and provisions of the Stockholm Convention.

6.2. Forgotten countries

As earlier mentioned, no studies could be found concerning the occurrence of PCBs, PBDEs and PFAS in Brunei and Timor-Leste nor any information on their consumption in these two countries. According to PCB inventory conducted in 2016-2017, 119 out of 592 inventoried transformers contained PCBs at concentration higher than the 50-ppm threshold set by the Stockholm Convention and there remained more than 319 tons of PCB-containing oil in Myanmar (UNEP, 2020). Moreover, in 2016, Myanmar imported more than 56,000 liters of firefighting foam, only 2.4% of which were used and the remains stayed in stockage. Nevertheless, there is no record of scientific article working on the presence of PCBs, PBDEs and PFAS in Myanmar's aquatic environment. Myanmar, formerly known as Burma, has experience prolonged periods of military rule and political unrest (Stokke et al., 2022). The most recent military coup d'état, which took place in February 2021, further destabilized the nation, sparking widespread armed conflicts and resistance as well as a humanitarian crisis (Andrews, 2022; Manurung, 2021). Hence, the political turmoil within Myanmar may have hindered and limited the monitoring activities despite the potential risks of contamination from POPs, which have been mentioned during the POPs inventory and the national implementation plan for the Stockholm Convention published in 2020 (UNEP, 2020). For the case of Timor-Leste, the country obtained its independence and was recognized as a sovereign nation by the United Nations only 23 years ago, on May 20, 2002. As a newly independent nation, though Timor Leste understands the importance of environmental monitoring, it has not yet to develop a fully structured environmental management system including legislation, regulation, and adequately resourced institutions (World Bank, 2009). This knowledge gap should be due to the lack of environmental monitoring strategies linked to limited governmental and

population concern and capacity of local facilities (e.g., analytical laboratories). Despite being the signatory of the Stockholm Convention since 2002, Brunei has never ratified the Convention and has no national restriction on the production and usage of POPs (Sharkey et al., 2020 . Effectively, no National Implementation Plan of Brunei has been submitted to the Stockholm Convention.

Gong et al. (2018) reported however the long-range transport of organochlorine compounds (including PCBs) by air masses coming from Myanmar, inferring thus the atmospheric contamination in this country. These three countries are drained by several large river systems (the Irrawaddy River and the Salween in Myanmar, the Belait River and Tutong River in Brunei, and the Loes River and Laclo River in Timor-Leste) and are well connected to other Southeast Asian countries via maritime routes. This implies that the dissemination of POPs by water ways are highly possible. Given their participation and commitment to the Stockholm Convention, it is required to conduct the industrial POPs inventories and monitoring in Brunei, Myanmar and Timor Leste, especially in aquatic compartments. To facilitate the international cooperation on environment in the region, the Association of Southeast Asian Nations (ASEAN) has been making efforts by establishing ASEAN Environmental Cooperation Framework. The Association hosts biannually the ASEAN Ministerial Meeting on Environment (AMME) assisted by the ASEAN Senior Officials on Environment (ASOEN) with specific working groups to cover all priority areas in environmental cooperation.

6.3. Chlorinated paraffins, an orphan broad group of industrial POPs

Similarly to the three previously reviewed groups of contaminants, the chlorinated paraffins (CPs) are a POPs-like group of compounds, estimated to gather more than 30,000 homologues and isomers (South et al., 2022). CPs are complex mixtures of polychlorinated alkanes with different degrees of chlorination and the chlorine content varying from 30% to 72% (w/w). Based on the length of the carbon chains, CPs are classified into three groups: short-chain CPs (SCCPs, C10 -13), medium-chain CPs (MCCPs, C14 – 17) and long-chain CPs (LCCPs, > C18) having 18 or more carbon atoms (Bayen et al., 2006). Since CPs have high chemical stability, flame retardancy and low volatility, they have been used as additives in metal cutting fluids, plasticizers, flame retardants in rubber, paints, and textiles (Huang et al., 2020; van Mourik et al., 2015). It was estimated that during the period from 1930 to 2020, 32.5 million tons of CPs were produced, of which

28% were SCCPs (Mu et al., 2023). The global cumulative emissions of CPs by 2020 have reached an alarming figure of 5.2 million metric tons (C. Chen et al., 2022). Nevertheless, CPs can be carcinogenic and SCCPs were categorized in Group 2B as possibly carcinogenic to humans from the International Agency for Research on Cancer. CPs can induce several other toxicities, disrupt endocrine systems, and cause developmental problems (S. Chen et al., 2023). SCCPs have been considered as POPs by the Stockholm Convention when they were listed in Annex A (Elimination) in 2017. SCCPs have persistent, bio-accumulable and toxic (PBT) and very persistent, very bio-accumulable (vPvB) properties and they were added to the Candidate List of substances of very high concern for Authorization under REACH Regulation. Furthermore, MCCPs are now being under consideration for the listing under the Convention (UNEP, 2021a) were added to the Candidate List of Substances of Very High Concern (SVHC) under the REACH Regulation in 2021 and are planned to be added to the ROHS restricted substances list (Commission, 2022; ROHS Directive 2011/65/EU for electronic devices).

In national implementation plans submitted to the Stockholm Convention by the Southeast Asian nations, little information was reported on the inventory of CPs. Indonesia recorded an imported amount of 13,521 tons of CPs between 2014 and 2016 and suggested that Thailand was an exporting country with the cumulative amount of 1143 tons (UNEP, 2021b). However, Indonesia, Thailand and Vietnam have taken important steps by initiating plans to include SCCPs in their national inventories and regulatory frameworks, as indicated in their implementation plans.

Despite their utmost concern in term of toxicity and production tonnage, this huge group of POPs could not be added to our review article, because there were no scientific article reporting the presence of CPs in the aquatic environment of any of the 11 South-East Asian countries. Nonetheless, the presence of CPs in the region has been recorded in other environmental samples as well as in human serum samples collected in China, Korea and Japan (van Mourik, Toms, et al., 2020). Thus, McGrath et al. (2023) investigated the presence of CPs in indoor dust in Thailand and three other countries (Japan, Australia, and Colombia). In general, the concentrations of CPs in samples from Thailand were higher than those from other three countries with the median values being 29 $\mu g\ g^{-1}$ (SCCPs), 120 $\mu g\ g^{-1}$ (MCCPs) and 12 $\mu g\ g^{-1}$ (LCCPs). Moreover, CPs were detected in ambient air in Vietnam, China, and Australia (C. He et al., 2023). While the concentrations of SCCPs were

comparable between air samples collected in Vietnam and China, the levels of MCCPs were two times lower in samples from Vietnam. The vicinity of China and India, which are both the two biggest CP producers in the world, the increase in the production rate of MCCPs in China (452,000 tons compared to 236,400 tons of SCCPs produced in 2019, (S. Chen et al., 2023) and CP detection in air samples in the region, are all arguments in favor of the relevance of including these molecules in the regular environmental monitoring strategies of South-East Asia, besides PCBs, PBDEs and PFAS.

6.4. Analytical challenges

The industrial group of POPs on which this review article focuses gather, in the case of PCBs or PBDEs, 209 congeners, but can amount to thousands of molecules in the case of PFAS or CPs. In both latter cases, given i) the huge number of compounds identified, ii) the intrinsic limitations of the target-based approach to dozens of molecules, iii) and the unavailability of analytical standards, alternative analytical strategies for measuring a total content have recently been developed.

Among them, for PFAS, the Total Oxidizable Precursors Assay (TOPA) accounting for PFAAs precursors such as Fluorotelomer sulfonic acids (FTS), the Extractable Organically bound Fluorine (EOF), and the Non-Target Screening (NTS) represent the current trend in environmental monitoring of PFAS (Bugsel et al., 2023; Houtz & Sedlak, 2012; Simon et al., 2023). But, few laboratories have their analysis in hand worldwide.

Fifteen years ago, the analysis of CPs in environmental matrices was qualified as “the ultimate challenge of the analytical chemist” by Tomy, as these complex mixtures (i) cannot be separated by chromatography, (ii) cannot be classically quantified by using analytical standards, and their signal can be interfered by a lot of chlorinated pollutants (Tomy, 2009). Since then, many sophisticated analytical methods have been applied to resolve this inextricable issue and were recently reviewed for environmental and food matrices (van Mourik et al., 2018; X. Yu et al., 2022; Yuan et al., 2019). Currently, the method most widely accepted by the scientific community, even if it is only partially satisfactory, relies on the latest generation of instruments and a complex quantification method based on recording the signal from mixtures with different degrees of chlorination (van Mourik, Lava, et al., 2020). This analytical sophistication is undoubtedly one of the reasons for the lack of data generated in South-East Asia as well as the paucity

of data generated worldwide, in comparison with PBDEs, which were produced, used and regulated at fairly similar times and purposes. Moreover, the compliance with the actual regulation trend requires analytical optimization for the accurate determination of MCCPs (Schinkel et al., 2018). The development of a simple and rapid qualitative method (presence/absence) could constitute a first step and a sustainable approach for the elucidation of this group of compounds in developing countries.

On the other hand, owing to their physico-chemical properties, POPs are prone to accumulate in sediment and biota and are rather present at trace levels in water. This explains why very few data are available on fresh or marine waters, with the exception of PFAS, some of which being quite polar. However, for bio-accessibility and toxicity purpose, the dissolved fraction of pollutants can be more relevant. But this is hampered by the ultra-trace concentrations of hydrophobic organic compounds in the dissolved phase, the fleeting nature of aqueous discharges, and the problems of glass sorption and sample preservation, all the more so when the field can be a long way from the analysis laboratory. To overcome these limitations for POPs, the development and progress of water passive sampling has emerged over the last ten years as a powerful tool, having been the subject of methodological guidelines and interlaboratory validation trials for PCB and PBDE congeners with silicone and low-density polyethylene (Booij et al., 2024; Smedes et al., 2012; Vrana et al., 2015). In addition, very recently, analytical developments were proposed for the water passive sampling of SCCPs (Godéré et al., 2021, 2022) and regulated PFAS (Fang et al., 2021; Kaltenberg et al., 2022).

However, none of the data collected in the South-Asia waters over even the most recent period is the result of passive sampling. Yet thanks to high polymer-water partition coefficients, the equivalent volume of water sampled can range up to thousands of liters (Vrana et al., 2020). These tools provide a time-weighted average concentration of dissolved POPs, reducing the frequency of spot sampling needed to achieve reliable analysis, and the pre-concentration of POPs directly *in situ*, enabling to achieve the sensitivity requirement (LOQs) (Godéré et al., 2022). The advantage also lies in the fact that flows and risks can be assessed. It would therefore be judicious to apply these tools, which have already been widely used in other parts of the world for POPs, to better estimate the dissolved phase fraction of POPs in these regions.

7. Conclusions

Among the compounds listed under the SC, the 209 congeners of PCBS and PBDEs, as well as PFAS and CPs form four groups of industrial POPs comprising a vast number of molecules. Their worldwide environmental dispersion, the rapid industrialization and economic development in Southeast Asian countries as well as their proximity to major producers such as China and India and the massive importation of the contaminated products in the past, have heightened the vulnerability of ASEAN environments to such persistent pollutants. The differences, both in the phases of economic development and in the history of practices and rulings, implies specific trends of POP concentrations.

Over the past three decades, 121 research papers were addressing the occurrence of PCBs, PBDEs, and PFAS in the fresh and marine hydro-systems (water, sediment, and aquatic biota) across eight Southeast Asian countries. It shows that these countries are paying increasing attention since 2004, to document the quality status of the water resources, particularly in connection with the implementation of regulations, or with the edible resources. The summarized data provides in-depth information for the national policymakers to align with the objectives of the SC. But the efforts need to be strengthened. Indeed, this review suggests that despite the national and international regulations, the concentrations of these POPs in environment has shown no sign of decline for the past 35 years. The review also underscores the urgent need for the regular environmental monitoring of these broad groups of POPs, points out the knowledge gaps in some countries, such as Brunei, Myanmar and Timor Leste, and the data lack concerning chlorinated paraffins. The development of sum or qualitative methods and the implementation of passive sampling technologies are advised as future analytical developments, to help in intensifying the monitoring.

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1.5. Objectives of the doctoral research

Under the legislative and economic context presented above, the general objective of this thesis is to strengthen current knowledge on the occurrence and behaviors of PAHs, PCBs, PBDEs, CPs, and PFAS in the Vietnamese aquatic environment, and more precisely in the Red River located in North Vietnam. In addition, to the specific objective of reviewing the current state of knowledge concerning the studied compounds in Southeast Asian aquatic environment (publication N°1) and presented in this first chapter, the operational objective of my research work was to address the gaps on the contamination situation of the Red River by investigating the occurrence and the dynamic of five groups of organic contaminants in this hydro-system. This operational objective was broken down and addressed as follows:

1) Sub-objective 1: Extending spatial and temporal coverage of PAH measurements, providing refined source identification, and evaluating seasonal variations as well as potential associated risks, as discussed in chapter three (publication N°2);
2) Sub-objective 2: Addressing PCBs, PBDEs, and CPs in Red River sediment by generating original datasets, identifying possible contamination sources and examining how these compounds behave in relation to sediment characteristics, anthropogenic pressures, seasonal changes and regulatory impact, as presented in chapter four (publication N°3);
3) Sub-objective 3: Adapting and validating extraction method based on the US-EPA1633 and applying both targeted analysis and group-level quantification (i.e., total oxidizable precursors assay, TOPA), in both water and sediment of the Red River, for investigating a broad range of PFAS, even the portion of “hidden” PFAS that may not have been covered by the target approach, as detailed in chapter five.

Finally, the thesis closes with a conclusion section with perspectives and future research direction

To achieve these operational objectives, four sampling campaigns were carried out in the Red River (200 km transect from the city center of Hanoi to the Ba Lat estuary) from November 2022 to September 2023. Several extraction and analytical methods have been employed for the analysis of five groups of organic contaminants, as overviewed in chapter two and described in “Materials and Methods” section of each following chapter.

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The four sampling campaigns were carried out in the Red River, Vietnam with logistical and institutional support from the International Joint Laboratory (LMI) LOTUS, which includes promoting research aquatic systems in Southeast Asia. The samples pre-treatment was carried out in the laboratory of the department of Chemical and Environmental Technology, USTH.

The PhD work was divided into two main phases. The first half of the project was conducted at the LCE, Aix-en-Provence, where the analyses of PAHs, PCBs, PBDEs, and CPs were performed. The second half of the thesis took place at LEGOS, in Toulouse, where the analyses of per- and polyfluoroalkyl substances (PFASs) were carried out.

2. Materials and Methods

This chapter provides an overview of the study site, the sampling campaigns, and the analyses that were conducted on the various samples. It is then supplemented by a presentation of the materials and methods specific to each chapter of results and by information on the target analytes and the performance of analytical methods provided in summary tables in the Annex.

2.1. Study area and sampling campaigns

2.1.1. Study area

The Red River constitutes one of the major river systems in Southeast Asia. With a total length of 1126 km, the river originates in Yunnan Province, China, and flows through northern Vietnam before emptying into the Gulf of Tonkin (Dang et al., 2010; Nguyen et al., 2024). The Red River forms a large delta system, the Red River Delta in Northern Vietnam (*Figure 17*), after receiving its two tributaries, the Da and Lo Rivers, near the province of Viet Tri. From there, the river flows southeastward through Hanoi, the country's capital, developing into a complex network of distributaries before reaching the coast.

The delta spans about 14,300 km^2 and lies mostly below three meters in elevation (Luu et al., 2010), with a population of 24 million (General Statistics Office of Vietnam, 2024). The Red River delta is the second largest agricultural area (after the Mekong delta), dominated by rice cultivation (*Figure 18*), which accounts for about 66% of land use (T. P. Q. Le et al., 2022).

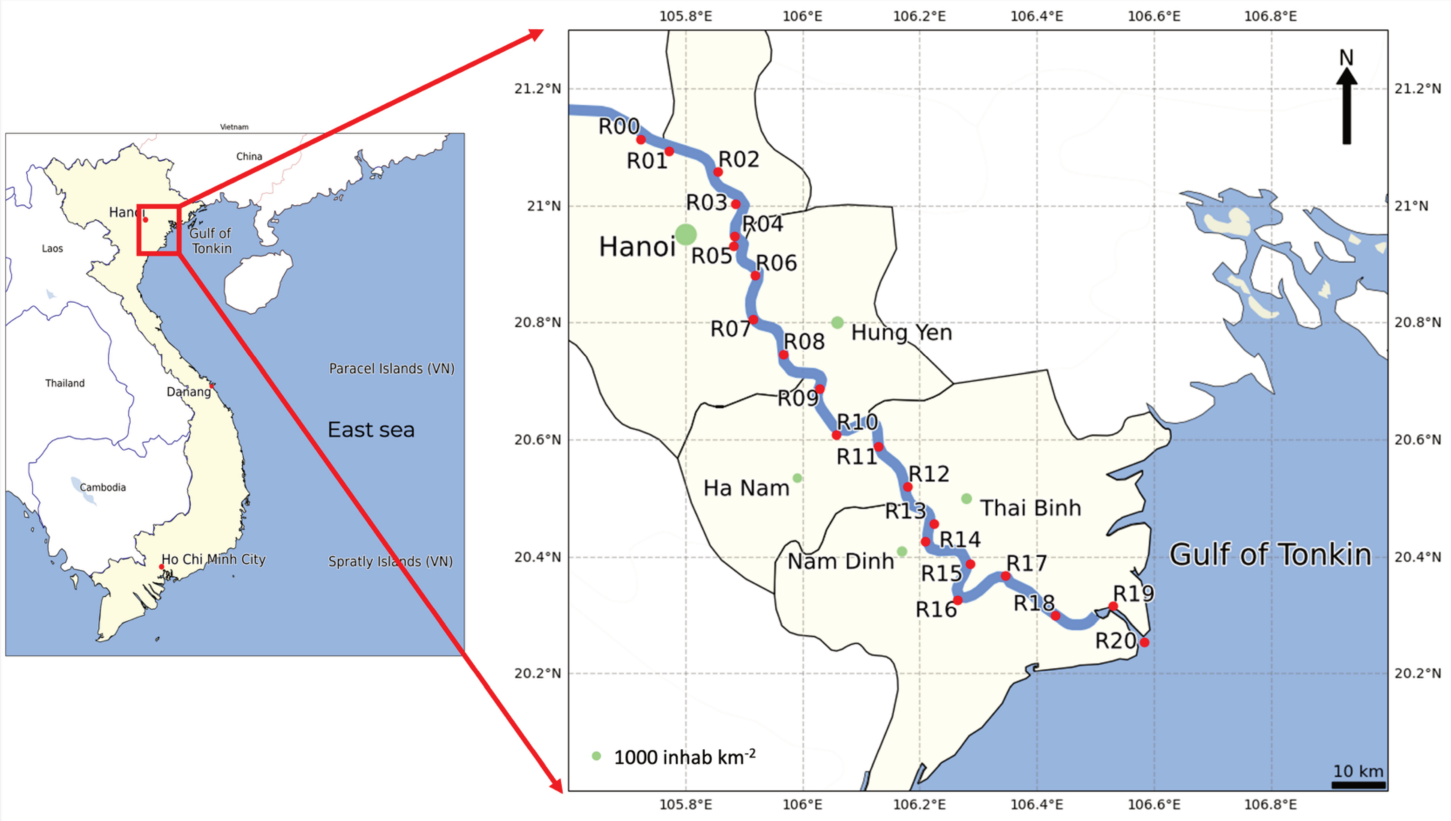

Figure 17: The Red River located in the northern Vietnam, forming the Red River delta and flowing pass the capital city of Hanoi.

The region is under the tropical monsoon climate with two distinct seasons: the **rainy season** from May to October and the **dry season** from November to April (Vinh et al., 2014). The hydrology of the Red River is strongly influenced by seasonal rainfall. The river has an average annual discharge of 2500 $m^3 s^{-1}$ (T. P. Q. Le et al., 2022), with the average flow rate ranging from about 1900 $m^3 s^{-1}$ in the dry season to 3100 $m^3 s^{-1}$ in the rainy season. The main branch of the Red River accounts for approximately 22.5% of the total water discharge (27 $km^3 yr^{-1}$) and 22.3% of the total sediment transport (7.8 $km^3 yr^{-1}$) to the Gulf of Tonkin. The river is a vital resource for agriculture, industry, and transportation, supporting the livelihoods of millions of people in the delta.

The main branch flows through Hanoi, the capital city, before entering the Gulf of Tonkin via the Ba Lat estuary. Hanoi, with the population of over 8.6 million and a density of 2585 inhabitants per km^2 (according to General Statistics Office of Vietnam, 2024) , is not only a political and cultural center but also a rapidly developing industrial hub of the country. Various industrial zones are located around the city, hosting diverse manufacturing industries such as textiles, chemical production, and food processing.

Downstream of Hanoi, the Red River traverses the rural zones with extensive agricultural practices and through other urban centers in the delta, e.g., Nam Dinh city with well-established textile industry. The combined impacts of population growth, intensive agricultural practices, and rapid industrial development highlight the vulnerability of the Red River to the pollution of persistent organic pollutants.

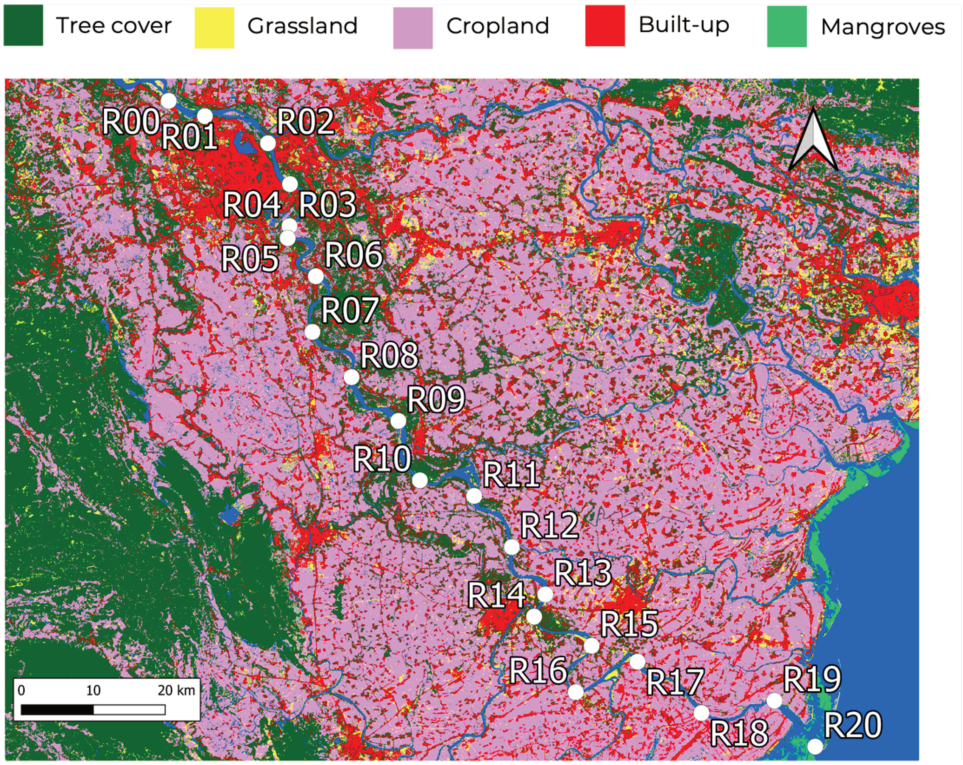

Figure 18: The land use map of the Red River delta and 21 sampling stations (the map was based on the layer provided by European Space Agency WorldCover project (2021))

2.1.2. Sampling campaigns

To study the occurrence of organic pollutants in the Red River, four sampling campaigns were carried out in the Red River in November 2022, March, June and September 2023. Twenty-one stations (named R00 to R20, *Figure 17 and Annex 8*) were chosen for sampling along the Red River, from Hanoi city center to the Ba Lat Estuary (Gulf of Tonkin). Sampling stations were systematically positioned at 10-kilometer intervals, except for station R04 at the discharge point of the Yen So Wastewater Treatment Plant. The summary of sampling campaigns, samling stations, and analyzed contaminants are presented in *Table 8*

Table 8: Overview of all samples collected during four sampling campaigns in 2022 and 2023, including sample types (water and sediment) and the contaminants analyzed in each campaign

	November 2022						March 2023					
	PAHs	PCBs	PBDEs	CPs	PFAS		PAHs	PCBs	PBDEs	CPs	PFAS	
	Sediment				Sediment	Water	Sediment				Sediment	Water
R00		✓	✓	✓	✓		✓				✓	✓
R01		✓	✓	✓	✓		✓				✓	
R02		✓	✓	✓	✓		✓				✓	✓
R03		✓	✓	✓	✓		✓				✓	✓
R04		✓	✓	✓	✓	✓	✓				✓	✓
R05		✓	✓	✓			✓					
R06		✓	✓	✓	✓	✓	✓				✓	✓
R07		✓	✓	✓			✓					
R08		✓	✓	✓			✓					
R09		✓	✓	✓	✓	✓	✓					✓
R10		✓	✓	✓	✓	✓	✓					
R11		✓	✓	✓			✓					
R12		✓	✓	✓			✓					
R13		✓	✓	✓	✓	✓	✓				✓	
R14		✓	✓	✓	✓	✓	✓				✓	
R15		✓	✓	✓	✓		✓				✓	✓
R16		✓	✓	✓			✓					
R17		✓	✓	✓			✓					
R18		✓	✓	✓	✓		✓				✓	✓
R19		✓	✓	✓			✓					
R20		✓	✓	✓	✓		✓				✓	✓

Table 8 (continued)

	June 2023						September 2023					
	PAHs	PCBs	PBDEs	CPs	PFAS		PAHs	PCBs	PBDEs	CPs	PFAS	
	Sediment				Sediment	Water	Sediment				Sediment	Water
R00	✓	✓	✓	✓	✓		✓				✓	
R01	✓	✓	✓	✓	✓	✓	✓				✓	
R02	✓	✓	✓	✓	✓	✓	✓				✓	
R03	✓	✓	✓	✓	✓	✓	✓				✓	
R04	✓	✓	✓	✓	✓		✓				✓	
R05	✓	✓	✓	✓			✓					
R06	✓	✓	✓	✓	✓	✓	✓				✓	
R07	✓	✓	✓	✓			✓					
R08	✓	✓	✓	✓			✓					
R09	✓	✓	✓	✓		✓	✓					
R10	✓	✓	✓	✓			✓					
R11	✓	✓	✓	✓			✓					
R12	✓	✓	✓	✓			✓					
R13	✓	✓	✓	✓	✓		✓				✓	
R14	✓	✓	✓	✓	✓	✓	✓				✓	
R15	✓	✓	✓	✓	✓		✓				✓	
R16	✓	✓	✓	✓			✓					
R17	✓	✓	✓	✓			✓					
R18	✓	✓	✓	✓	✓	✓	✓				✓	
R19	✓	✓	✓	✓			✓					
R20	✓	✓	✓	✓	✓	✓	✓				✓	

2.2. Extraction methods

The overview of the extraction and analytical protocols for five groups of contaminants of interest are presented in *Figure 19*.

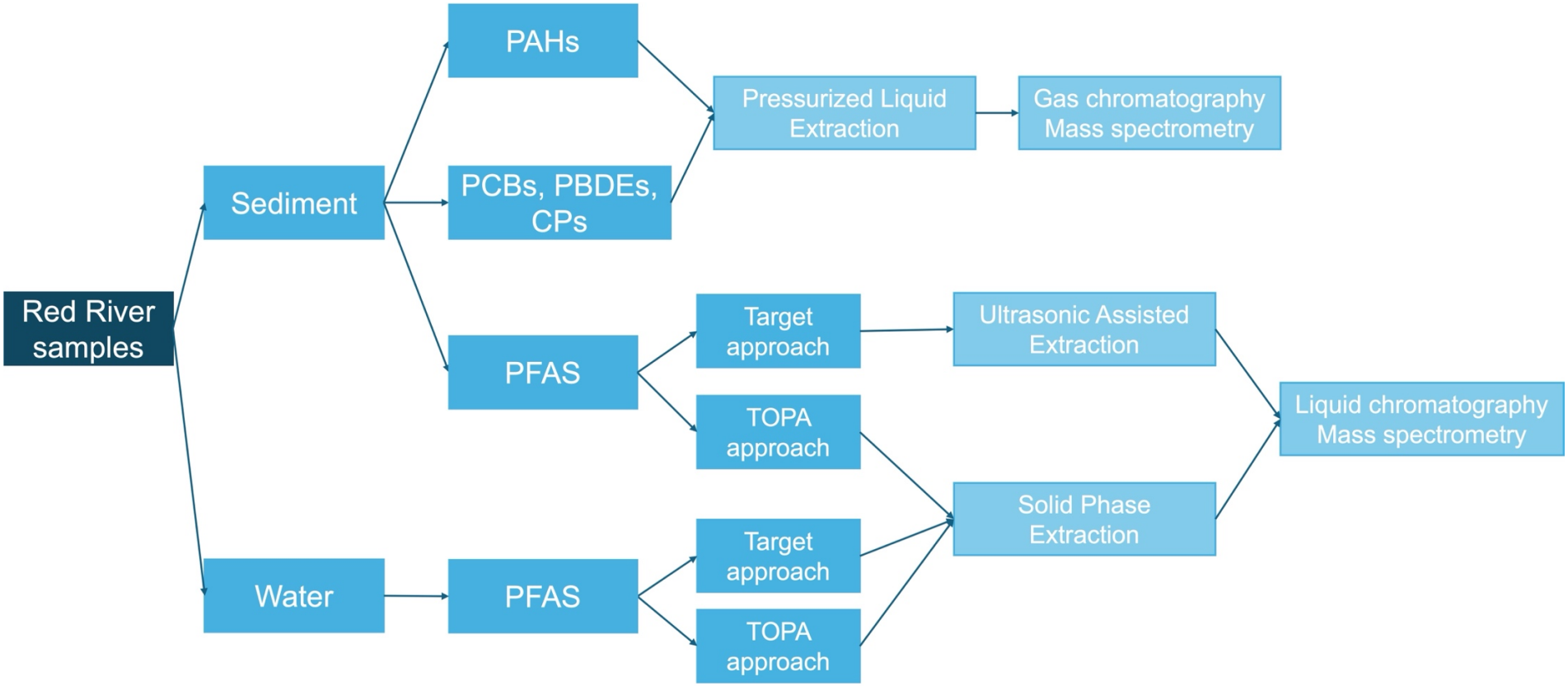

Figure 19: General scheme presenting the extraction and analytical methods for sediment and water samples for PAHs, PCBs, PBDEs, CPs, and PFAS.

2.2.1. Sediment samples

The sediment samples were sampled using a Van Veen grab. Approximately the upper 5 cm of the surface sediment in the riverbed were collected and put into 500-mL pre-combusted aluminum boxes (500 ˚C for 8 hours). The samples were stored in the freezer (-80 ˚C) and later freeze-dried for 5 days using a Buchi Lyovapor L-200. Then the dry samples were sieved to a size class inferior to 200 µm using a stainless-steel mesh and stored in pre-combusted aluminum boxes prior to further analysis and extraction.

Total organic carbon (TOC) of the 200-µm sieved sediment samples was derived from the total carbon and total inorganic carbon contents, which were determined by Multi N/C 2100 analyzer from Analytik Jena. The TOC values were obtained from the difference between the total carbon(TC) and total inorganic carbon (TIC). A Mastersizer 3000 (Malvern Instrument Ltd., Panalytical, Malvern, UK), equipped with an automated wet dispersion unit (Hydro LV), was used to determine sediment particle size via laser diffraction analysis (LDA). TOC contents and particle size distribution of sediment samples from four sampling campaigns are presented in *Annex 8*.

2.2.1.1. PAHs

The list of 16 priority PAHs is presented in *Annex 1*.

PAHs were extracted from sediment samples using Pressurized Liquid Extraction (ASE 200 Accelerated Solvent Extractor, Thermo Scientific™ Dionex™, USA) with 33 mL stainless-steel extraction cells. A glass fiber filter was first inserted, followed by a mixture of 10 g of freeze-dried sediments and Fontainebleau sand. A phenanthrene-d_{10} solution (50 ng $sample^{-1}$) was added as a deuterated surrogate standard. The extraction was performed using a Hexane/Dichloromethane (Hex/DCM, 1:1, v/v) solvent mixture, with two static cycles of 5 minutes each at 150 °C under 100 bar pressure. After each cycle, the extract was flushed into a collection vial and evaporated until 1 mL afterwards.

A cleanup step was carried out using Resprep EPH Silica cartridges (6 mL, 2 g). The samples were loaded, followed by the passage of 6 mL of Hex for cleanup and 6 mL of DCM

for PAHs elution. The eluate was then evaporated under a gentle air stream and reconstituted in 200 µL of Hex in micro-vials. Before analysis, a Chrysene-d_{12} solution (50 ng sample^{-1}) was added as an internal standard.

2.2.1.2. PCBs, PBDEs, and CPs

Seven PCBs congeners (*Annex 2*), namely PCB-28, -52, -101, -118, -138, -158, and -180, and the eight PBDE congeners (BDE-28, -47, -99, -100, -153, -154, -183, and -209, *Annex 3*) were chosen as target analytes. The quantification of SCCPs is performed with the certified mixture Chloroparaffin (C10-13, 63% of chlorine by mass).

The extraction of PCBs, PBDEs and SCCPs were adapted from Godéré et al. (2022) and Rigal et al. (2024) with slight modifications. The extraction was performed using Pressurized Liquid Extraction (ASE 200 Accelerated Solvent Extractor, Thermo Scientific™ Dionex™, USA) with 33 mL stainless-steel extraction cells. Ten mg of sediment sample was spiked with surrogate standards (20 ng sample^{-1} of d_5-PCB30, d_3-PCB156, BDE 79, and BDE 189). The extraction process used the mixture of Hex/DCM (1:1, v/v) as solvent and underwent two cycles of 7 minutes each at the temperature of 100 and the pressure of 100 bar. The extracts were then concentrated under a gentle air flow until 1 mL and went to the clean-up step on silica gel cartridges (SPE Supelclean™ 154 LC-Si, 1 g, 6 mL, Supelco). The extract (1 mL) was loaded into the cartridge pre-conditioned with 6 mL each of DCM, mixture of Hex/DCM (1:1, v/v), and Hex. After that, the cartridge was eluted with 8 mL of Hex for Fraction 1 (F1, containing PCB and BDE) and 8 mL of Hex/DCM (1:1, v/v) for Fraction 2 (F2, containing CPs). Each fraction was thereafter evaporated under a gentle air flow and reconstituted in 200 µL of Hex. 20 ng sample^{-1} of d_5-PCB 116 and BDE 140 were added to F1 and 20 ng sample^{-1} of $^{13}C_6$-hexachlorobenzene were added to F2 as internal standards.

2.2.1.3. PFAS

Fifty-three PFAS have been chosen for analysis in this PhD. thesis. They are perfluoroalkyl carboxylic acids (PFCAs), perfluoroalkyl sulfonic acid (PFSAs), perfluoroalkyl sulfonamides, poly- and perfluoroether carboxylic acids (PFECAs), fluorotelomer sulfonic acids (FTS), Ether sulfonic acids, Fluorotelomer carboxylic acids (FTCAs), and polyfluoroalkyl phosphate diesters.

The detailed information of targeted PFAS is presented in *Annex 4*.

The extraction methods for PFAS are illustrated in *Annex 14*.

2.2.1.3.1. Target approach

Sediment samples (1 g, dw) were spiked with surrogate standards (solution MPFAC-HIF-ES at 2.5 – 50 ng sample^{-1}, *Annex 12*). The samples are then extracted with 5 mL of 0.3% NH_4OH in MeOH by ultrasonication for 30 min. After centrifugation at 3500 rpm for 10 min, the supernatant was transferred to a 50 mL PP tube. The extraction step was repeated two more times, each lasted for 10 min. The combined extract was then treated with 10 mg of activated carbon (ENVI Carb), mixed for 5 min, and centrifuged at 3500 rpm for 10 min. The supernatant was transferred and diluted to 250 mL with UPW. After being pH-adjusted, the sample was passed through the Oasis WAX (Waters™, 150 mg, 6 cc, particle size: 30 µm) cartridges pre-conditioned with 4 mL fractions of 1% ammonium hydroxide (NH_4OH) in methanol (MeOH), MeOH and ultrapure water (UPW). After the sample loading, the cartridges were washed with 4 mL UPW and vacuum-dried for 5 minutes. Next, the cartridges were eluted using 5 mL of 1% NH_4OH in MeOH. The extracts were then evaporated under gentle flow of N_2 gas until the volume of 500 µL. After the addition of internal standards (solution MPFAC-HIF-IS at 2.5 – 10 ng sample^{-1}, *Annex 13*), the extracts were filtered using nylon filter (17 mm diameter, 0.2 µm pore size) and transferred into injection vials.

2.2.1.3.2. Total oxidizable precursor assay (TOPA)

The main principle and the oxidation process are described in the introduction of Chapter 5. TOPA approach for sediment samples were adapted from (Göckener, Fliedner, et al., 2022; Houtz et al., 2013). Sediment samples (100 mg, dw), 4.76 g of $Na_2S_2O_8$ (200 mM) and 2 g of NaOH (500 mM) and 100 mL of UPW were added into the glass bottle and incubated at 85 °C overnight. The samples were then collected and centrifuged at 3500 rpm for 10 min and later diluted with 100 mL UPW to obtain the final volume of 200 mL. After being pH-adjusted and spiked with the surrogate standards (solution MPFAC-HIF-ES at 2.5 – 50 ng sample^{-1}, *Annex 12*), the SPE protocol described in section 2.2.1.3.1 was performed.

2.2.2. Water samples – PFAS

Water samples were collected using a stainless-steel collector and transferred into pre-combusted (500 °C, 8 hours) 1-L glass bottles. The samples were filtered by glass fiber filter (GF/F, 0.7 μm, Whatman™) and stored in freezer at -80 °C before extraction. A total of 23 water samples were chosen to be analyzed: 6 from November 2022, 9 from March 2023, and 8 from June 2023.

2.2.2.1. Target approach

Surrogate standards (solution MPFAC-HIF-ES at 2.5 – 50 ng sample^{-1}, *Annex 12*) were added to the samples. Water samples (250 mL) were passed through Oasis WAX cartridges as described in section 2.2.1.3.1.

2.2.2.2. TOPA approach

The TOPA procedure was adapted from (Houtz & Sedlak, 2012). 250 mL of filtered water sample were transferred into a glass bottle. In each bottle, 4 g of $Na_2S_2O_8$ and 3.8 mL of NaOH 10 M were added as oxidizing agents. The bottles were placed in the oven at 85 °C overnight to promote oxidation. Afterwards, the samples were cooled down to room temperature, and the pH was then adjusted between 5-9 with HCl solution (37%). The samples were subsequently subjected to solid phase extraction using Oasis WAX cartridges following the protocol described in Section 2.2.1.3.1.

2.3. Analytical methods

2.3.1. Sediment particle size and Total Organic Carbon (TOC)

The size distribution of sediment particles was determined by Mastersizer 3000 with Hydro LV wet sampler dispersion from Malvern Panalytical. Total organic carbon (TOC) content (%) of the 200-μm sieved sediment samples was derived from the total carbon and total inorganic carbon contents, which were determined by Multi N/C 2100 analyzer from Analytik Jena.

2.3.2. Gas chromatography – Mass spectrometry

2.3.2.1. PAHs

PAHs were analyzed using capillary gas chromatography coupled with mass spectrometry (GC–MS; Autosystem XL GC and TurboMassTM- from Perkin Elmer, USA). The chromatographic and mass spectrometry conditions were adapted from the literature (Kanzari et al., 2014) with slight modifications. Analytes were separated on an Elite-5MS Capillary Column (30 m × 0.25 mm I.D. × 0.25 µm, Perkin Elmer) in spitless injection mode. The injector was programmed from 50 °C (0.1 min isothermal) to 250 °C (200 °C min^{-1}). The GC oven was temperature-programmed from 40 °C (2 min isothermal) to 120 °C (45 °C min^{-1}), then to 295 °C (5 °C min^{-1}) and held isothermally for 20 min. The mass spectrometer was operated under positive electron impact with the ionization energy of 70 eV with the source temperature set at 240 °C. The data acquisition was done simultaneously both in Full scan and Single Ion Monitoring (SIM) mode. The list of quantifier ions is presented in *Annex 9*.

2.3.2.2. PCBs, PBDEs, and CPs

PCBs and CPs were measured by a gas chromatography (GC-7890A, Agilent Technologies) coupled with tandem mass spectrometry system (MS-3000A, Agilent Technologies) with electron ionization (GC/EI-MS/MS). The analysis of PBDEs was performed using gas chromatography (GC Clarus 600 AutoSystems XL) in combination with electron capture negative ion mass spectrometry (GC/ECNI-MS, MS Clarus 600C, PerkinElmer). The list of quantifier ions is presented in *Annex 10* The analysis conditions were adapted from the literature (Godéré et al., 2022, Rigal et al., 2024) and detailed in *Annex 15.*

2.3.3. Liquid chromatography – Mass spectrometry

The analysis of PFAS was performed by reverse phase liquid chromatography coupled with high-resolution mass spectrometry (THERMO, Orbitrap Exploris 120). The separation was achieved on DIONEX AcclaimTM C18 (2.1 mm x 150 mm x 3 µm) column

at 40 °C. The injection volume was 10 µL and the total running time was 19 min. The mobile phase was 2 mM of ammonium acetate + 0.1% acetic acid in H2O (phase A) and 2 mM of ammonium acetate + 0.1% acetic acid in MeOH (phase B). The flow rate was kept constant at 0.4 mL min^{-1}. Elution was performed under gradient conditions: 0 min 95:5 A/B, 0.5 min 95:5 A/B, 3 min 50:50 A/B, 11 min 0:100 A/B, 16 min 0:100 A/B, 16.1 min 95:5 A/B. The list of quantifier and qualifier ions and MS conditions are presented in *Annex 11* and *Annex 16*, respectively.

2.4. Quality assurance and Quality control

All the containers of samples and instruments for extraction steps were thoroughly cleaned using MiliQ water, acetone, and methanol. The glassware was rinsed with acetone and calcinated at 350 ^{o}C for 8 hours. Procedural blanks were performed along extraction batches to verify the possible contamination. Detailed information on blank samples could be found in the section of "Material and Methods" in each chapter. Real water and sediment samples were systematically spiked with surrogate standards as indicated in section 2.2 of this chapter for the monitoring the performance of extraction methods. Results were corrected with the recovery rate of the surrogates and values of the blank samples. The LOD values for PAHs are shown in *Annex 9*, for PCBs, PBDEs, and CPs in *Annex 10*, and PFAS in *Annex 11*.

Calibration points for quality control were injected regularly during the injection processes. The accuracies are presented in the "Quality assurance and Quality control" section in each chapter.

3. Polycyclic Aromatic Hydrocarbons

Context and objective

The literature review has provided a general overview of the occurrence of long-term organic contaminants in aquatic environments in Vietnam. PAHs are ubiquitous environmental pollutants that originate from both natural processes and, more importantly from anthropogenic sources. Their hydrophobic nature favors their accumulation in sediments, where they may persist over long time periods and pose ecological risks for organisms and, through bioaccumulation, to higher trophic levels and human. Chapter 1 has shown that, in spite of the recognition of PAHs as hazardous substances and their inclusion in the Vietnamese sediment quality guidelines, existing data on their occurrence in sediments across the country remains fragmentary. Most of the available studies have focused on highly urbanized or industrialized zones, such as the samples in two major metropoles of Hanoi and Ho Chi Minh City, or coastal zones with bustling marine transports. These studies consistently reported elevated concentrations of PAHs associated with pyrogenic inputs, reflecting the dominance of combustion-derived sources in rapidly urbanizing zones. However, knowledge is scarce when it comes to major river systems, especially the Red River, which plays a vital role in supplying water, supporting fisheries and inland transports across one of the most densely populated and economically dynamic regions of Vietnam. The monitoring in the Red River has so far been limited to localized measurements and assessments.

Chapter 3 is intended to address this knowledge gap by presenting a study on PAHs in the sediments of the Red River. This chapter fulfills the sub-objective 1 outlined in section 1.5, which are to quantify the concentrations of PAHs in sediments, to assess their spatial and temporal variations, and to identify potential sources and associated ecological risks.

The study area covers a 200 km transect of the Red River, extending from the city center of Hanoi, through the deltaic region with intensive agricultural activities, and reaching the Ba Lat Estuary at the Gulf of Tonkin. A total of 21 sediment samples were

collected during three sampling campaigns conducted in March 2023 (late dry season), June 2023 (early rainy season), and September 2023 (late rainy season). Sediment extraction was performed using pressurized liquid extraction, and the samples were later analyzed by gas chromatography coupled with mass spectrometry to quantify 16 targeted PAHs listed as priority pollutants by the United States Environment Protection Agency (ATSDR, 1995).

Results' overview

The concentrations of the 16 priority PAHs (∑16PAHs) in the Red River sediments were generally low, with maximum values slightly above 100 ng g^{-1} dw. These concentrations fall within the lower range of values reported worldwide for riverine sediments. This suggests that, despite the increasing anthropogenic activities along the Red River, the overall contamination of PAHs is relatively low

Clear seasonal patterns were observed in the dataset. The mean ∑16PAHs during the rainy season was significantly higher compared to those measured in the dry season. This trend indicates the hydrological dynamics of the river system, where enhanced runoff during the rainy season mobilizes and transports PAHs accumulated from the surrounding areas into the river. The temporal variation also showed that concentrations during the early rainy season (June 2023) were higher than those in the late rainy season (September 2023). This phenomenon can be explained by the "first flush" effect, which implies that the discharge from the initial rainfall events after a prolonged dry period tend to carry greater amounts of accumulated contaminants, leading to elevated contaminant inputs in the river system.

In terms of spatial distribution, the highest concentrations of PAHs were detected in stations located close to urban and residential zones, particularly in areas subject to dense human activity and heavy traffic. These findings emphasize the influence of local emission sources, where rapid urbanization, transportation, and domestic discharges exert substantial pressure on the river system. On the contrary, lower concentrations were observed in less densely populated and peri-urban zones, and in the Ba Lat estuary with highly preserved Xuan Thuy National Park.

Source apportionment based diagnostic ratios and principal component analysis (PCA) revealed that pyrogenic inputs dominates the PAH burden in Red River sediments.

Biomass and fossil fuel combustion, largely related to transportation emissions, thermal power plants, and traditional agricultural practices, presented as the main contributors. As of ecological and human health risks, the concentrations observed in the Red River sediments were generally below guidelines thresholds. The calculated indices suggests that PAHs in the study area exert low carcinogenic and mutagenic risks, with values complying with Vietnamese sediment quality standards. Concerning the ecological risk assessment, the presence of PAHs may rarely generate risks on sediment quality and aquatic organisms.

The detailed results and discussions are included in the following publication N°2 entitled "Spatial and Seasonal Variation, Source, and Risk Assessment of Sedimentary Polycyclic Aromatic Hydrocarbons from the Red River to the Ba Lat Estuary (Vietnam)" published in *Marine Pollution Bulletin*.

Partial conclusions

By examining samples collected across multiple sites and campaigns, the study has provided a detailed characterization of PAHs in one of Vietnam's most important river systems.

Overall, the findings confirm that PAHs remain a relevant class of legacy contaminants in the Red River, despite their long-recognized toxicity and inclusion in sediment quality guidelines. The results illustrate not only the ongoing presence of these pollutants but also their usefulness as indicators of anthropogenic pressures in rapidly urbanizing and industrializing regions. By filling a critical data gap, this chapter contributes to a more comprehensive understanding of sediment contamination in Vietnam and provides a scientific basis for future monitoring and management strategies in Southeast Asia.

Publication N°2: **Spatial and Seasonal Variation, Source, and Risk Assessment of Sedimentary Polycyclic Aromatic Hydrocarbons from the Red River to the Ba Lat Estuary (Vietnam)**

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Abstract:

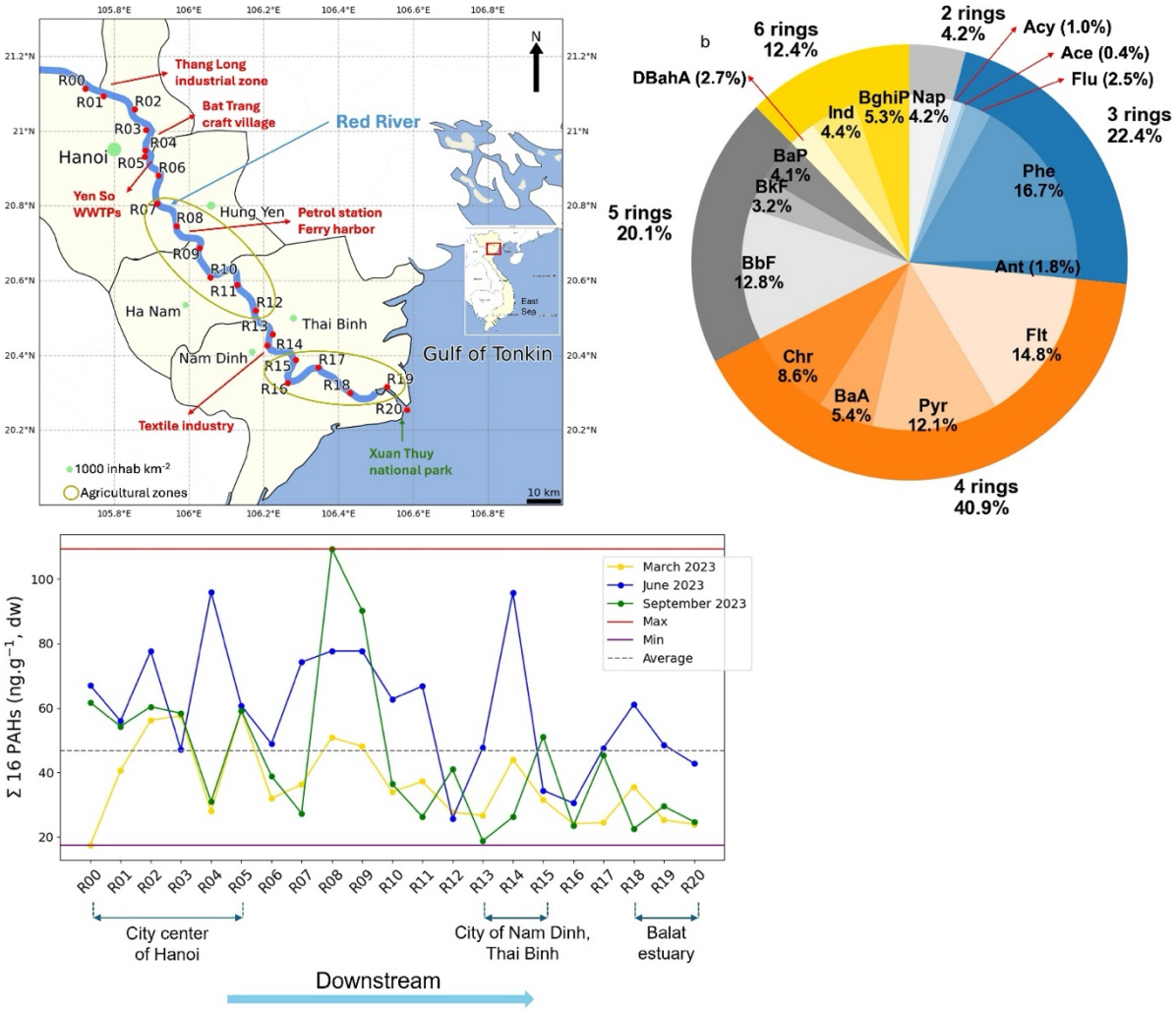
The spatial-seasonal variation, source diagnostics, and ecological risks of 16 priority polycyclic aromatic hydrocarbons (PAHs) were examined in surface sediments of the Red River, from the capital city of Hanoi to the Ba Lat Estuary (Northern Vietnam). The average sum concentrations of the 16 PAHs in 21 sediment samples collected in March, June, and September 2023 were 35.0±12.2 ng g^{-1} dw, 56.0±19.7 ng g^{-1} dw, and 43.3±23.3 ng g^{-1} dw, respectively. The highest concentrations of PAHs were measured in sampling stations close to city centers and residential areas, where road traffic and domestic activities are prevalent. The PAH levels were significantly higher during the early rainy season (June) compared to the dry season (March). The composition of the 16 PAHs followed the same pattern for the three sets of samples: 4-ring > 5-ring ~ 3-ring > 6-ring >2-ring. Diagnostic ratios suggested that PAHs at the three periods were mainly from pyrogenic sources, coming both from the combustion of biomass and fossil fuels. According to international sediment quality guidelines, the levels of PAHs quantified in the Red River would present minimal ecological risk to living organisms. As far as it concerns, this is the first study on the contamination of PAHs in the Red River, from Hanoi city center to Ba Lat Estuary in the Gulf of Tonkin.

Keywords: PAHs, Sediment, Gulf of Tonkin**,** pyrogenic sources, risk assessment, environmental quality standards.

Highlights:

- Σ16PAHs in the Red River sediments are in the low range of those reported worldwide.
- Mean Σ16PAHs in the rainy season was significantly higher than in the dry season.
- The highest concentrations were measured in urban and residential areas.
- Combustion of biomass and fossil fuel were the main sources of PAHs.
- PAHs exert low carcinogenic/mutagenic risks, compiling with national guidelines.

Graphical abstract:



3.1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a group of organic compounds containing two or more fused benzene rings. They are ubiquitous in the environment, being emitted naturally (forest fires, volcanic eruptions) (Vergnoux et al., 2011) or through anthropogenic activities (Kishida et al., 2007; Saha et al., 2009). There are two main categories of PAHs depending on their origin: 1) petrogenic PAHs, which are produced by the low-temperature transformation of organic matter. These are mainly low molecular weight (LMW) PAHs, i.e., with 2-3 rings. They mainly come from crude oil and its derivatives. 2) pyrogenic PAHs, which are released following incomplete combustion

at high temperature of fossil fuels (oil, diesel, gasoline, coal, etc.) and biomass. These are mainly high molecular weight (HMW) PAHs, i.e., ≥ 4-rings (Kishida et al., 2007; Pradhap et al., 2021; Saha et al., 2009). PAHs have been detected in various environmental compartments, including air and dust (Castel et al., 2023a; Larsen & Baker, 2003; Pham et al., 2019; Quoc Anh et al., 2019), water and sediment (Asia et al., 2009; Hoang et al., 2021a; Mbusnum et al., 2020; Xu et al., 2007) and soils (Boll et al., 2008; Hoa et al., 2020)

The chemical stabilization of PAHs is achieved by the delocalization of electrons of the π-bonds in the benzene ring, which made them stable and persistent (Souza et al., 2018). Due to their low volatility and water solubility, and their hydrophobic nature, PAHs tend to adsorb onto particulate organic matter. Thus, in the aquatic environment, they preferentially accumulate in sediments, where they can persist for periods ranging from several months to years (Iwegbue et al., 2021; Zhao et al., 2020); but also in living organisms and food chains (Al-Battawi et al., 2024; Pham et al., 2020). The intake of PAHs via oral absorption, inhalation and dermal interactions poses significant risks to the human health (Ambade et al., 2021). For instance, PAHs have been detected in urinary samples taken in several Asian countries (Y. Guo et al., 2013), in blood serum of adults in Malaysia (Al-Battawi et al., 2024), and in human milk collected in Croatia (Mendaš et al., 2024). Being mutagenic, carcinogenic and endocrine disrupting, PAHs are categorized as legacy organic contaminants, 16 of which (*Annex 9*) had been designated as priority pollutants by the United States Environmental Protection Agency (US-EPA) since the 1970s. (ATSDR, 1995; Hoang et al., 2021a).

Rapid growth of urbanization and industrialization contributes significantly to the emission of PAHs into the environment (X. Li et al., 2022). Vietnam, a developing country in Southeast Asia, has witnessed a fast rate of industrialization, significantly contributing to the rapid development of the nation's economy. According to the Vietnamese General Statistics Office (GSO), the country's gross domestic product (GDP) had increased by 7.1% in 2024. The manufacturing industries (e.g., textiles and footwear, electronics and machinery) contributed to 26.3% of Vietnam's GDP in 2023 (Asian Development Bank, https://kidb.adb.org, accessed February 2025). The Red River Delta in northern Vietnam is one of the most dynamic economic regions in the country. Red River is the principal river branch in the delta and the second largest river in Vietnam. It originates in the mountains in Dali (Yunan Province, China) and enters Vietnam territory at Lao Cai

Province before passing through Vietnam's capital city of Hanoi and flowing to the Gulf of Tonkin (part of the East Sea) at Ba Lat estuary. The downstream section of the Red River (i.e., Red River Delta) is under the influence of Southeast Asia's sub-tropical monsoon climate that is divided into a rainy season (from May to September) and a dry season (from October to April, Vinh et al., 2014). As the largest river in the Vietnamese northern region, the Red River plays a vital role as a freshwater source for drinking and irrigation water, and waterways for transportation. Rice cultivation, livestock production, aquaculture, and the manufacturing of textiles and construction materials are among the most prominent economic activities in the vicinity of the river (Dang & Tran, 2020; Tran et al., 2021). The latter passes through several major cities and provinces in the Red River Delta: Hanoi (2,594 inhab km^{-2}), Hung Yen (1,417 inhab km^{-2}), Thai Binh (1,193 inhab km^{-2}), Nam Dinh (1,135 inhab km^{-2}), and Ha Nam (1,037 inhab km^{-2}) (data in 2024 by GSO, https://www.gso.gov.vn/, accessed in September 2025). Hence the river is under strong anthropogenic influence and susceptible to the contamination of chemicals, particularly PAHs.

The occurrence of PAHs has already been reported in the region of the Red River Delta (Duong et al., 2014; Hoa et al., 2020; Hoang et al., 2021a; Hung et al., 2015; Toan et al., 2020). For example, the 16 priority PAHs were detected in sediment samples from urban rivers in Hanoi and Haiphong (Duong et al., 2014; Hoang, Takahashi, Da Le, Hoang, et al., 2021; Toan et al., 2020), in sewage sludge (Hung et al., 2015) or in soil and sediment from e-waste recycling sites in Hung Yen province (Hoa et al., 2020). On a national scale, the presence of PAHs has also been mentioned in atmospheric particulate matters and dust (Guigue et al., 2025; Quoc Anh et al., 2019; Thanh Trung et al., 2021), and in bivalves (Thuy et al., 2018). However, the presence of PAHs has not been thoroughly investigated in the Red River itself, despite its importance and vulnerability. The aim of this study was to document the occurrence, molecular distribution and associated risks of PAHs in surface sediment collected in the downstream part of the Red River, from Hanoi to the Ba Lat Estuary. To evaluate the spatial and seasonal variations, we investigated the concentrations of the 16 priority PAHs, on 21 sampling stations along the Red River, collected during the dry (March 2023) and rainy season (June and September 2023).

3.2. Materials and Methods

3.2.1. Study area and sampling

The Red River, with a total length of over 1100 km, originates in Yunnan Province, China, and flows through northern Vietnam before discharging into the Gulf of Tonkin (Dang et al., 2010; Nguyen & Tran, 2024). The river forms the Red River Delta. Covering an area of approximately 14000 km^2, the delta supports a population of about 24 million (General Statistics Office of Vietnam, 2024). It represents the country's second largest agricultural region after the Mekong Delta, with rice cultivation as the dominant land use, accounting for around 66% of the delta's area (Le et al., 2022a). The region is characterized by a tropical monsoon climate with two distinct seasons: a rainy season from May to October and a dry season from November to April (Vinh et al., 2014). River hydrology is strongly driven by seasonal rainfall, with an average annual rainfall of 1600 mm and annual discharge of 2500 $m^3 s^{-1}$ (Dang et al., 2010; Le et al., 2022a).

This research focused on the downstream section of the Red River passing through major cities of Hanoi, Nam Dinh, Ha Nam, Hung Yen, and Thai Binh in the Red River delta. Twenty-one samples (numbered R00 to R20, *Figure 20*) were collected from the upstream outskirts of Hanoi to the Ba Lat estuary in three different sampling campaigns in 2023: March (late-dry season), June (early-rainy season) and September (late-rainy season). The detailed information on the sampling stations is presented in *Annex 8.* Stations R00 to R03 are located in Hanoi, the capital city of Vietnam with a population exceeding 8 million in 2023. Stations R04 and R05 are at the discharge point of the Yen So wastewater treatment plant. Stations R08 and R09 are positioned near a harbor. Stations R13 and R14 are in Nam Dinh city, in close proximity to Thai Binh city. The remaining stations are located in suburban and rural areas.

The superficial sediment samples were collected in the riverbed using a Van Veen Grab Sampler. Approximately 500 g of surface sediments were taken and stored at -20 °C in pre-calcinated aluminum containers. In the laboratory, the samples were freeze-dried using a Freeze Dryer (Buchi, pro200, Switzerland) and sieved with a mesh size of 200 μm. They were, then, stored in aluminum foil at room temperature prior to extraction and analysis. The mesh size of 200 μm was chosen because hydrophobic organic contaminants

preferentially bind to fine particles that have higher specific surface area and organic matter content, while coarse grains typically contain negligible contaminant loads and may dilute measured concentration (Cui et al., 2011; Talley et al., 2002). This sieving size has been adopted in previous studies (Kanzari et al., 2014; Näslund et al., 2008).



Figure 20: The Red River and the 21 sampling stations (Red River Delta, northern Vietnam). Cities with high population density, industrial area, and agricultural zones are highlighted.

3.2.2. Chemical reagents

The mixture of the 16 PAHs (PAH mixture 933 at 20 µg mL^{-1} in dichloromethane) was purchased from LGC Standards, Ltd. Deuterated surrogate and internal standard, phenanthrene-d_{10} and chrysene-d_{12}, were purchased from Dr. Ehrenstorfer Laboratories.

Dichloromethane (DCM for HPLC, ≥ 99.8%) and hexane (Hex for HPLC, ≥ 97%) were obtained from Sigma Aldrich

3.2.3. Extraction of PAHs from sediment samples

The extraction of PAHs from sediment samples was performed using pressurized liquid extraction (ASE 200 Accelerated Solvent Extractor, Thermo Scientific™ Dionex™, USA) equipped with 33 mL stainless-steel extraction cells. After inserting a glass fiber filter, the cell was filled with a mix of 10 g of freeze-dried sediments and Fontainebleau sand. Phenanthrene-d10 solution (50 ng $sample^{-1}$) was added to the extraction cell as a deuterated surrogate standard. Each sample was extracted with a mixture of Hex/DCM 1:1 (v/v) and two cycles of 5 min in static mode at 150 °C under the pressure of 100 bar were programmed. After extraction and extract reconcentration to 1 mL, a cleanup step was performed on Resprep EPH Silica cartridges (6 mL, 2 g) which were conditioned by passing 3 mL of DCM and then 6 mL of Hex. Samples were loaded, then 6 mL of Hex were passed through as a cleanup and then and 6 mL of DCM were used for elution of PAHs. The eluate was evaporated under a gentle air stream and reconstituted in 200 µL of Hex in micro-vials. Prior to analysis, chrysene-d_{12} solution was added to the micro-vials as an internal standard (50 ng $sample^{-1}$).

3.2.4. Analytical methods

Total organic carbon (TOC) of the 200-µm sieved sediment samples was derived from the total carbon and total inorganic carbon contents, which were determined by Multi N/C 2100 analyzer from Analytik Jena. The TOC values were obtained from the difference between the total carbon (TC) and total inorganic carbon (TIC). TC was quantified by combusting 700 mg of sediment at 950 ˚C with CO2 detection by pre-calibrated (using CaCO3, 12% C) non-dispersive infrared (NDIR) analyzer. TIC was determined from 500 mg of sediment which were reacted with 2 mL of phosphoric acid (H_3PO_4, 40%) at 80 °C under agitation (350 rpm), and CO2 was measured by the same NDIR system.

A Mastersizer 3000 (Malvern Instrument Ltd., Panalytical, Malvern, UK), equipped with an automated wet dispersion unit (Hydro LV), was used to determine sediment particle size via Laser Diffraction Analysis (LDA). Reverse osmosis water (ROW) served as the dispersant, initially introduced at 500 mL to establish the background measurement.

Sediment was then gradually added until the obscuration level reached between 0.1 and 20% (Sedláčková et al., 2024). The measurement range was set from 0.01 to 3500 µm, with both background and sample measurements conducted over 10 s and repeated five times. A refractive index of 1.457 was applied, reflecting the dominance of sand (SiO_2) in the sediment composition. The particle size of sediment was classified as clay (<4 µm), silt (between 4 and 63 µm) and sand (> 63 µm) (Valentine, 2019).

The PAHs were analyzed using capillary gas chromatography coupled with mass spectrometry (GC–MS; Autosystem XL GC and TurboMass™- from Perkin Elmer, USA). The chromatographic and mass spectrometry conditions were adapted from literature (Kanzari et al., 2014) with slight modifications. Analytes were separated on an Elite-5MS Capillary Column (30 m × 0.25 mm I.D. × 0.25 µm, Perkin Elmer) with spitless injection. The injector was programmed from 50 °C (0.1 min isothermal) to 250 °C (200 ˚C min^{-1}). The GC oven was temperature-programmed from 40 °C (2 min isothermal) to 120 °C (rate: 45 ˚C min^{-1}), then to 295 ˚C (rate: 5 ˚C min^{-1}) and held isothermally for 20 min. The mass spectrometer was operated under positive electron impact with the ionization energy of 70eV with the source temperature set at 240 °C. The data acquisition was done simultaneously in both Full scan and Single Ion Monitoring (SIM) mode.

3.2.5. Quality assurance and quality control

The quality control procedures were strictly followed throughout the whole sampling and laboratory treatments and analyses. All the glassware was rinsed with acetone and oven-calcinated at 350 °C for 8 h to remove organic impurities. An eleven-point calibration curve (2, 5, 10, 25, 50, 100, 200, 250, 500, 750, 1000 ng mL^{-1}, R^2 = 0.99) was prepared for the quantification. Procedural blanks composed of Fontainebleau sand were carried out between each batch of 10 samples. Among the six blank samples, all 10 out of 16 PAHs were below the limit of detection (LOD). The exceptions were Nap detected in 5 blanks (averaged concentration 0.08 ng g^{-1} dw), Phe, Flt and Pyr each detected in 4 blanks (0.28, 0.21, and 0.51 ng g^{-1} dw, respectively), and Flu and Chr each detected in 2 blanks (0.12 and 0.05 ng g^{-1} dw, respectively). The reported concentrations are blank-corrected. The recovery of phenanthrene-d10 surrogate ranged from 51 to 113%. If the surrogate recovery was < 50%, the sample was reprocessed. The PAH concentrations were adjusted according to the surrogate recovery rate. The LOD for 16 PAHs at a signal-to-noise ratio

of 3 were from 0.01 – 0.09 ng g^{-1} dw, as presented in *Annex 9*. The extraction method was validated by analyzing certified reference material (CRM) as described elsewhere (Mbusnum et al., 2020). Fortified experiment was performed by spiking Fontainebleau blank sand with native and surrogate compounds to check the recovery rates of the extraction methods. The recovery rates ranged from 88±12% and 101±10%. Calibration points were added regularly during the injection for quality control. The accuracy of the method was between 84±9% and 108±7%.

3.2.6. Risk assessments

Among the 16 priority PAHs, when considering the carcinogenic potential to humans, 7 HMW PAHs (i.e., BaA, Chr, BbF, BkF, BaP, DBahA and Ind) are classified as carcinogenic PAHs (C-PAHs). To estimate the carcinogenicity of PAHs in mixture, the carcinogenic potency factor of other C-PAHs is expressed relatively to that of BaP (Castel et al., 2023b). The toxic equivalent quotient (TEQ) for carcinogenic PAHs is calculated as follows:

$$\sum TEQ_{C-PAHs} = \sum(C_i * TEF_i) \quad (1)$$

where C_i is the concentration of individual carcinogenic PAH and TEF_i is the corresponding Toxic Equivalent Factor.

Similarly, the mutagenic equivalent quotient (MEQ) for C-PAHs is calculated as follows:

$$\sum MEQ_{C-PAHs} = \sum(C_i * MEF_i) \quad (2)$$

where C_i is the concentration of individual carcinogenic PAH and MEF_i is the corresponding Mutagenic Equivalent Factor.

The TEF and MEF for each carcinogenic PAH are listed in *Annex 9.*

3.3. Results and discussion

3.3.1. Spatial and seasonal variation of PAH concentrations

The sum concentrations of the 16 priority PAHs (Σ16PAHs) were quantified, and their spatial-seasonal variations are shown in *Figure 21.* The individual concentrations of the

16 PAHs in the 21 sediment samples of the three sampling campaigns are shown in *Annex 17*. Σ16PAHs ranged from 16.2 to 57.9 ng g^{-1} dw in March, from 24.5 to 94.7 ng g^{-1} dw in June, and from 17.5 to 108.0 ng g^{-1} dw in September. The PAH contamination levels might be divided into 4 different levels based on the sum concentrations: low contamination (< 100 ng g^{-1}), moderate contamination (101 – 1000 ng g^{-1}), high contamination (1000 – 5000 ng g^{-1}) and very high contamination (>5000 ng g^{-1}) (Baumard et al., 1998). According to this classification, all samples exhibited low contamination levels, except station R8 in September, where the concentrations of 16 PAHs (108.0 ng g^{-1} dw) were in the moderate contamination criteria.

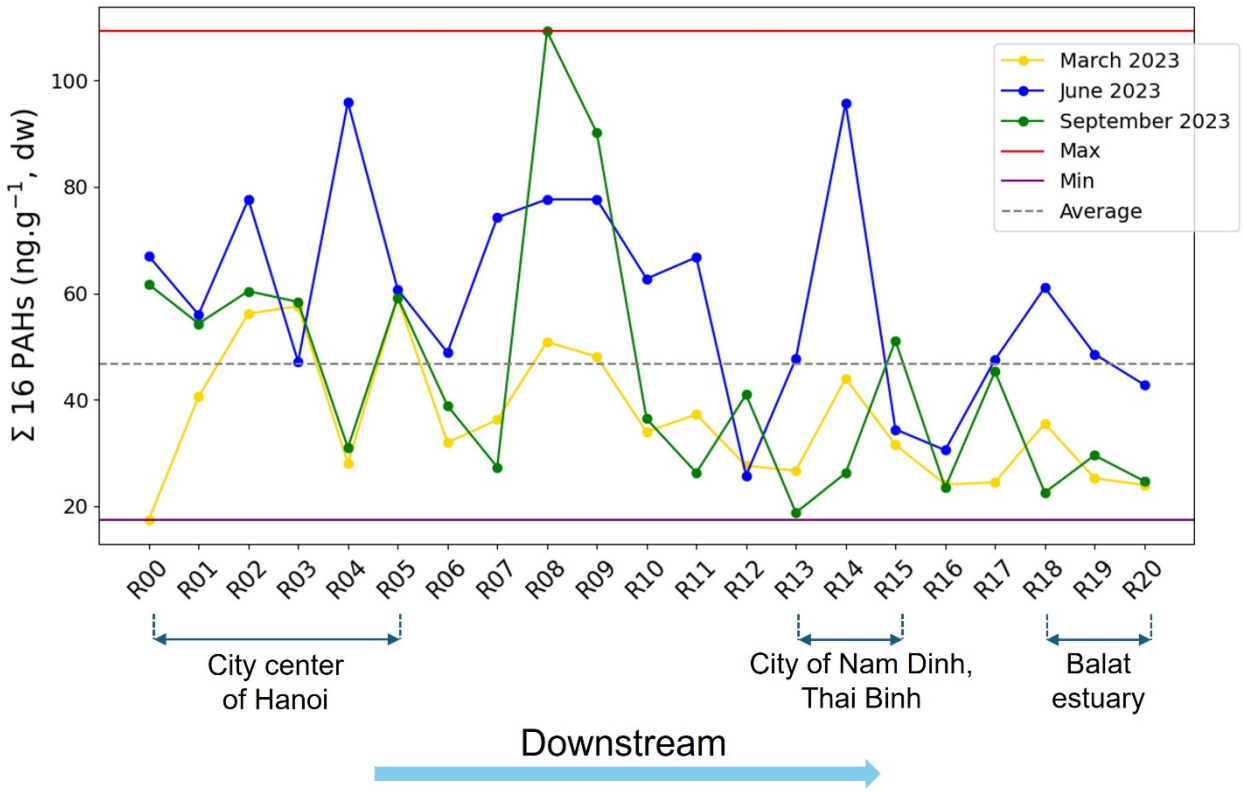


Figure 21: The sum concentrations of the 16 PAHs (Σ16PAHs) in the Red River in sediments from three sampling campaigns in 2023.

Additionally, the Vietnamese government established the newest National Technical Regulation on Sediment Quality (QCVN 43:2017/BTNMT) in 2017, setting the limit of individual 11 (out of 16) priority PAHs in both freshwater and marine sediment (*Table 9*). Accordingly, the PAH levels in Red River sediment samples collected in 2023 were well below the national standards.

Table 9: Vietnam's National technical regulation of PAHs on sediment quality (QCVN 43:2017/BTNMT).

Compounds	Freshwater sediment (ng g^{-1})	Marine sediment (ng g^{-1})
Nap	391	391
Acy	128	128
Ace	88.9	88.9
Flu	144	144
Phe	515	544
Ant	245	245
Flt	2355	1494
Pyr	875	1398
BaA	385	693
Chr	862	846
BbF	-	-
BkF	-	-
BaP	-	-
DBahA	135	135
Ind	-	-
BghiP	-	-

The contamination of PAHs has been proven to be highly related to the content of organic matter in the sediment (X. Li et al., 2022; Simpson et al., 1996; J. L. Zhou & Maskaoui, 2003). Being hydrophobic, PAHs have a tendency to accumulate into the particulate organic matter which may ultimately be deposited during sedimentation (Kishida et al., 2007; Lee et al., 2017). The TOC values in sediment samples from the three campaigns ranged 0.19 - 1.23% (detailed data is presented in *Annex 8*). Sediment composition was also analyzed, expressed as the proportions of sand, silt, and clay (detailed in *Annex 8*). In the Red River samples, sediment consisted of approximately 45% sand, 55% silt, and less than 0.1% clay. A Pearson's correlation test was performed to investigate the correlation between the TOC content and the concentrations of the 16 individual PAHs in the sediment samples. The test also included the sediment

composition, i.e., the percentage of clay, silt, and sand in the samples (*Annex 8*). The results are presented as correlation matrix and the p-value matrix for samples from March, June and September 2023 (*Annex 18, Annex 19, and Annex 20,* respectively). In general, the 16 PAHs showed a moderate to strong positive correlation with the organic content with a high level of statistical significance ($r = 0.3\text{-}0.8$, $n = 63$, $p < 0.01$), confirming that the hydrophobic PAHs tend to preferentially partition into organic matter in the sediment. The good correlation between the organic content and the PAH levels was also supported by previous works (Bajt, 2012; Hoang et al., 2021a; Souza et al., 2018). Additionally, the concentrations of individual PAHs were also positively correlated with each other, as well as with the content in silt and clay. In contrast, they showed strong negative correlations with the sand component in the sediment. These results reflect the strong influence of organic carbon content and granulometric properties of sediments on the overall distribution of sedimentary PAHs (Cao et al., 2024).

No clear increasing or decreasing trend in Σ16PAHs could be observed from the upstream to the Ba Lat estuary (*Annex 21)*. However, stations located in the city center of Hanoi, particularly from R00 and R05, exhibited elevated Σ16PAHs for all three campaigns. The highest concentrations were recorded at stations R04 and R05, which are near Hanoi's wastewater discharge points, in June and March, respectively. Similarly, stations R08 and R09, which are near a local ferry harbor and a gas station for boats, showed high Σ16PAHs in March and June, with a contamination spike in September (*Annex 21c*). Further downstream, elevated Σ16PAHs were observed at stations R13 and R14, in the vicinity of Nam Dinh city, one of the major urban areas in the Red River Delta, aside Hanoi. Notably, station 14 serves as the discharge point of the Kien Giang River, which is connected to Thai Binh City's canals and passes through residential areas. These areas experience frequent human activity, contributing to the high PAH load in sediments. In contrast, the lowest Σ16PAHs during the three campaigns were detected in stations (from R15 to R17) located in the rural or sub-urban residential zones. In addition, the estuary zone (stations R18, R19, and R20) was also recorded with low Σ16PAHs. These zones are less densely populated and, as a consequence, less impacted by the anthropogenic activities. Indeed, the estuarine zone includes Xuan Thuy National Park, announced in 1989 as the world's 50th and Vietnam's first Ramsar site by UNESCO. This protected area imposes strict limitations on anthropogenic activities to preserve habitats

of native and migratory species, particularly waterbirds. Consequently, restricted industrial and agricultural inputs likely contribute to the lower levels of contamination observed in this zone.

The seasonal variation of PAH concentrations in sediment among three sampling campaigns was investigated. As shown in *Figure 22*, it appeared that the mean ∑16PAHs of samples from June (early-rainy season) was significantly higher (t-test, $p < 0.05$, *Annex 22*, *Annex 24*) than those from March (late-dry season) and September (late-rainy season), while mean Σ16PAHs were not significantly different in March and September (t-test, $p > 0.05$, *Annex 23*).

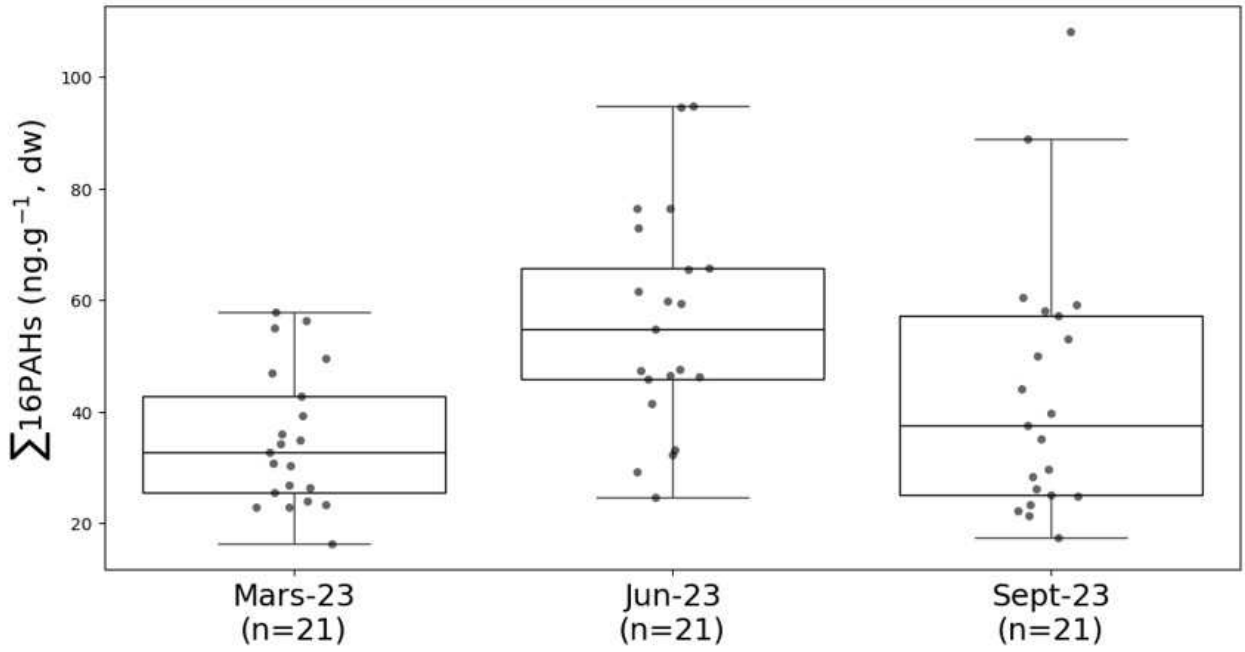


Figure 22: Distribution of Σ16PAHs in the Red River sediments in March, June, and September

Higher levels of sedimentary PAHs during the wet season than during the dry season have already been recorded in the literature (Babut et al., 2019; Hoang, Takahashi, Da Le, Hoang, et al., 2021; Thuy et al., 2021). During the rainy season, the runoff may wash off the particles previously contaminated with PAHs from the inland and deposit them in the riverbed during the sedimentation, ultimately increasing the concentration of PAHs in sediment relatively to the dry season. Interestingly, the mean Σ16 PAHs in September was lower than that in June ($p < 0.05$, *Annex 24*). Thuy et al. (2021) reported the decreasing trend in PAH levels in the estuarine and coastal sediments in Cangio wetland during the

rainy season. The Red River in June may have experienced the "first flush" effect as described by Maniquiz-Redillas et al. (2022). The "first flush" effect is a phenomenon where initial discharge of a runoff event contains significantly higher amounts of contaminants compared to later stages of the event. The authors further explained that long dry periods preceding the rainy season has allowed for significant accumulation of contaminants in terrestrial environments. With the arrival of precipitations in early rainy season, these accumulated contaminants (including PAHs) can be mobilized by surface runoff, enter the aquatic system and deposited in sediments. As the rainy season progresses, the intervals between rainfall events become shorter, reducing the time for pollutant build-up on land and consequently decreasing the amounts of terrestrial contaminants transported to the river. Besides, during the rainy season, the river discharge intensifies, potentially leading to the mobilization of the riverbed and resuspension of the contaminated sediment (Glaser et al., 2020; Montuori et al., 2022). Guigue et al. (2017) conducted a series of sediment resuspension experiments and observed a threefold increase in the concentration of 34 PAHs in the dissolved phase after 14 days. This increase indicates that PAHs were progressively released from sediment into the water column during resuspension, potentially reducing PAH concentrations within the sediment itself. Thus, the combined effects of the "first flush" at the beginning of the rainy season and sediment resuspension occurring throughout the season may help explain the higher PAH concentrations in the June samples than those collected in September.

Overall, the PAH concentrations measured here in the Red River sediment samples (16.2 – 108.0 ng g^{-1} dw) were substantially lower than those reported in previous studies conducted elsewhere in Vietnam. Kishida et al. (2007) analyzed sediments samples from urban lakes in Hanoi, lagoons in Hue, and urban and sub-urban channels and Saigon River in Ho Chi Minh City with the average concentration of 49 PAHs being 1600, 130, and 1900 ng g^{-1} dw, respectively. Kim Nguu river, an urban river which is located at the city center of Hanoi and received 1/3 of the city's wastewater, exhibited exceptional levels of PAHs in sediment, i.e., from 218 to 751 µg g^{-1} (Hung et al., 2015), which were three to four order of magnitude higher than those of the present study. Average PAH concentrations in sediment samples from urban rivers in Hanoi (2,594 inhab km^{-2}), Hai Phong (1,391 inhab km^{-2}), Danang (993 inhab km^{-2}) and Ho Chi Minh City (4,554 inhab km^{-2}) (data in 2024 by GSO, https://www.gso.gov.vn/, accessed in September 2025) were reported to be 2700,

500, 100 and 1500 ng g^{-1} dw respectively (Duong et al., 2014). Another recent work conducted by Babut et al. (2019) showed Σ16PAHs from 49 to more than 900 ng g^{-1} dw in the Saigon River. On the international scale, the PAH levels in Red River sediment were relatively low compared to other rivers in the neighboring countries, for example the Chao Phraya River in Thailand (mean concentration of 263 ng g^{-1} dw; Boonyatumanond et al., 2006); Kelanta River in Malaysia (169 – 461 ng g^{-1} dw; Vaezzadeh et al., 2021); Baisha and Moshui rivers in China (mean concentrations of 469.7 and 3979.5 ng g^{-1} dw, respectively; Han et al., 2021), and Yeongsan River and Nakdong River estuaries in South Korea (mean concentration of 340 and 380 ng g^{-1} dw, respectively, Lee et al., 2022).

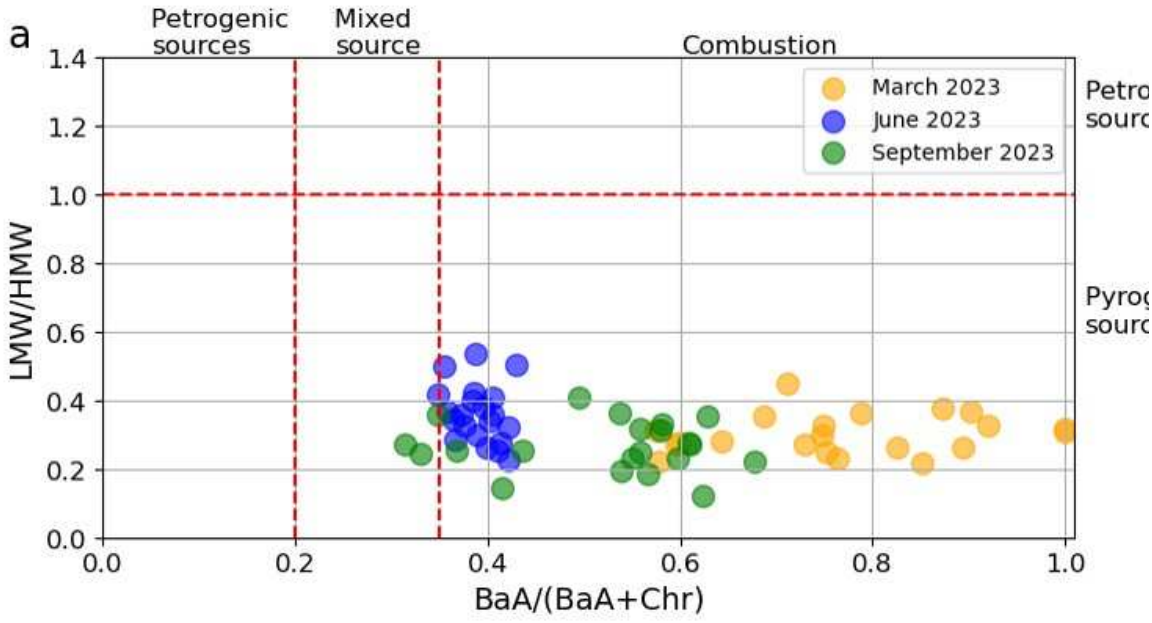
On the other hand, our results from the Red River were in agreement with those from the Poxim River, Brazil (2.2 – 28.4 ng g^{-1} dw; Souza et al., 2018), the coastal area of Hormozgan Province, Iran (14.54 - 84.00 ng g^{-1} dw; Abootalebi Jahromi et al., 2020) and one sample also collected in the Red River (33 ng g^{-1} dw; Hoang et al., 2021). Given the positive correlation with PAHs, low TOC content could be the explanation for the low PAH levels found in Red River sediment. The low PAH levels were coherent with the granulometric composition of the sediment. Clay, the finest fraction in sediment, favors the accumulation of organic carbon and organic contaminants (Liu et al., 2008; M. Singh et al., 2016). As presented in *Annex 8*, clay content was almost absent in the Red River sediments, less than 0.13% in all samples. This very low content supports the observation of low PAH concentrations. This was also the case for the Poxim River and Hormozgan Province whose TOC contents in sediments were relatively low and comparable to those in the Red River sediments, at 0.3 – 2.5% and 0.004 – 0.04%, respectively.

3.3.2. PAH composition and source identification

All 16 targeted hydrocarbons were found in all 21 samples collected in June. On the contrary, in March, while 13 out of 16 PAHs were detected in every sample, B(k)F, Nap and Chr had the respective detection frequencies (DF) of 10, 15, and 86%. Only B(a)P (DF = 90%) and DBahA (DF = 65%) were not found in all September samples. *Annex 25* shows the PAH profiles in sediments from the three sampling campaigns. The 16 PAHs were categorized into groups corresponding to their number of benzene rings (2-, 3-, 4-, 5-, and 6-ring PAHs).

The contribution of the 5 groups of PAHs in the sediments remained quite similar from the end of dry season to the end of the rainy season *Annex 25*. Overall, the 4-ring PAHs, which included Flt, Pyr, B(a)A and Chr, were the most abundant, making up of more than 40% of total PAHs detected in all three periods. They were followed by 5-ring PAHs (25.6% of total PAHs in March, 23.4% in June, and 23.7% in September) and 3-ring PAHs (22.7% in March, 22.3 % in June, and 20.2% in September).

To determine the main sources of PAHs, diagnostic ratios have been frequently used in previous studies *(Annex 26)*. Giuliani et al. (2008) suggested that the principles of the ratios were based on the thermodynamic stability of PAH congeners, the composition of contaminant sources and the changes in the relative abundances of PAHs between sources and sediment. As the LMW/HMW ratios for all samples from three sampling campaigns were well below 1 (*Figure 23a, Annex 27*), this indicated the pyrogenic source of PAHs (Hoa et al., 2020). The BaA/(BaA+Chr), Ind/(Ind+BghiP) and Flt/(Flt+Pyr) ratios supported the pyrogenic sources of PAHs. Given the ratio of Ind/(Ind+BghiP) > 0.2 and Flt/(Flt+Pyr) > 0.4, PAHs in sediments from March, June and September were derived both from fossil fuel combustion (engines, exhaust fume from vehicles and vessels) and burning of biomass (wood, grass and agricultural by-products) (*Figure 23b, c; Annex 27*).



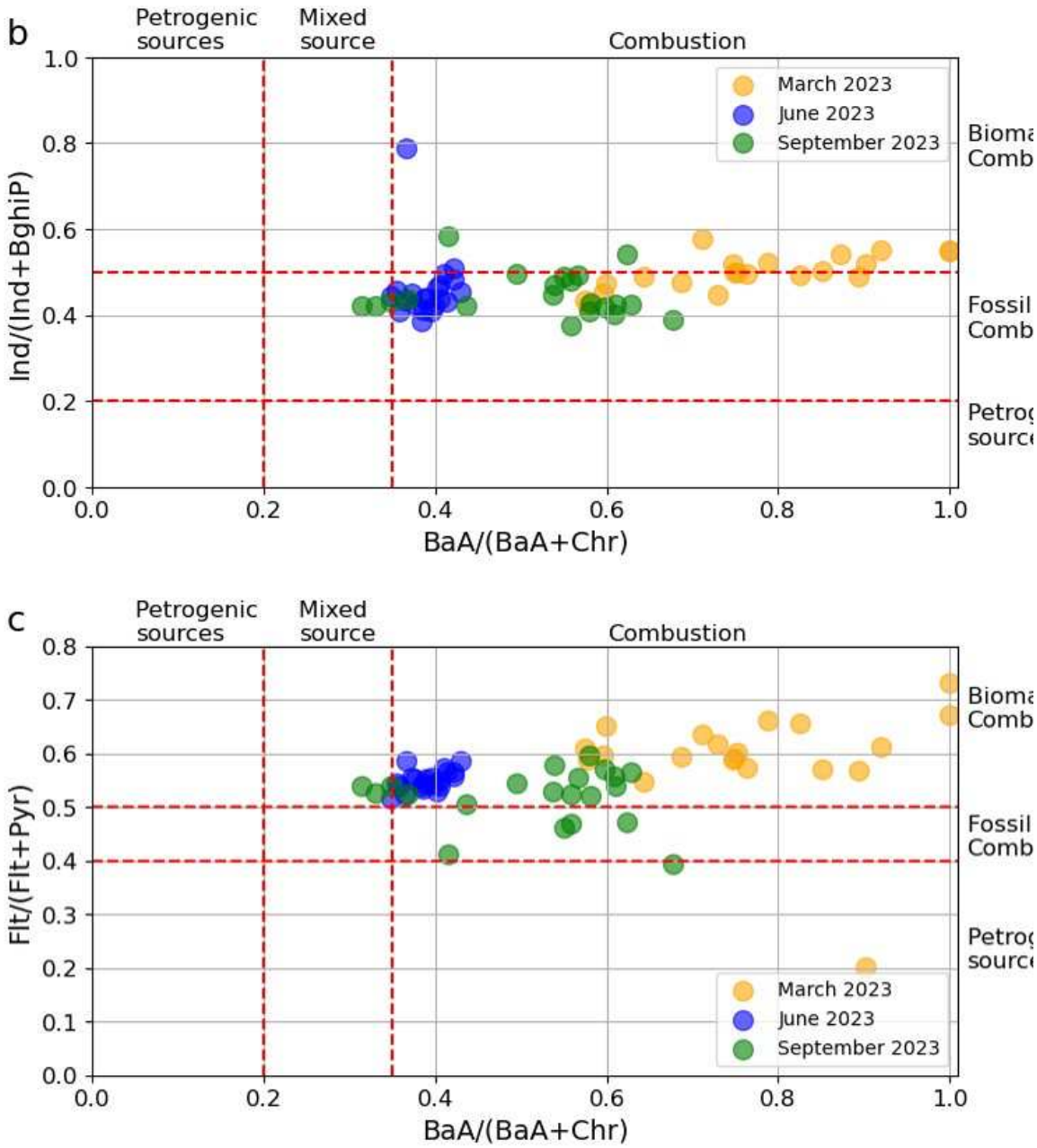


Figure 23: Plot of a) LMW/HMW, b) Ind/(Ind+BghiP) and c) Flt/(Flt+Pyr) vs BaA/(BaA+Chr) diagnostic ratios for March, June, and September samples.

Furthermore, a principal component analysis (PCA) was performed to identify the source of PAHs. The loadings of variables (∑16 PAHs, TOC, sediment composition of clay, silt, and sand) and the percentage of variance are presented in *Annex 28*. The detailed PCA results are presented in *Annex 29*. The two principal components (PC1 and PC2) were responsible for 67.6% of the total variance of samples in March, 77.9% in June, and 70.5% in September. PC1 was weighted towards HMW PAHs (Flt, Pyr, BaA, Chr, BbF, BaP, IP, and BghiP) for March samples. For June and September samples, the PC1 showed high loads

in Phe, Ant, Pyr, and Chr. While BghiP and BaP could be linked to motor vehicle emissions, Ant, Phe, Flt, Pyr, and Chr were associated with coal, wood, biomass, and incineration (Larsen & Baker, 2003; X. Zhang et al., 2024). In summary, the PCA results and their interpretation for the sediment samples from all three sampling campaigns correspond well with the conclusions drawn from diagnostic ratios. This indicates that both approaches are congruent and implies that PAHs in the Red River sediments originate primarily from pyrogenic sources.

Being the largest river in the northern region of Vietnam and flowing to the Gulf of Tonkin, the Red River is one of the most important waterways for transportation. Ferries and boats transporting merchandise and serving daily commutes and tourism purposes travel regularly along and across the river, emitting considerable amounts of exhaust engine fumes. In addition, bridges connecting the river's banks with high traffic volume could contribute to the emission of PAHs along the river. Moreover, the Red River delta is the second largest rice-producing area in Vietnam with two cultivating seasons. The most common method to dispose of agricultural wastes (e.g., rice straw) is direct open-site burning in the rice field. Even though it is very convenient for the farmers to burn waste after each crop, this unsustainable practice has significantly released contaminants into the surrounding environment, including PAHs (Le et al., 2022; Pham et al., 2019). Intense socioeconomic activities along the Red River could explain the pyrogenic sources of PAHs in the sediment samples, originating from the combustion of both fossil fuels and biomass.

3.3.3. Risk assessments

Sediment quality guidelines (SQG) were developed as effective tools to assess the ecological risks and to predict the toxicological effects of chemicals on biota, especially on benthic organisms. Long et al. (1995) proposed a guideline with two values: the Effect Range Low (ERL) and the Effect Range Median (ERM), respectively identified as the 10th and 50th percentile values of a series of data on the concentrations of chemicals and the corresponding ecotoxicological effects. This guideline has been widely used and adapted for the assessment of sediment (Ambade et al., 2022; Bajt, 2012; Kanzari et al., 2014; Mbusnum et al., 2020). If the chemical concentrations are below the ERL values, it means that the biological effects would rarely occur. When the concentrations fall in the possible-effect range between the ERL and ERM values, the effects would occur occasionally.

Finally, the negative effects would frequently occur if the concentrations were higher than the ERM value and lie within the probable-effect range.

In parallel, the Canadian Council of Ministers of the Environment (CCME) developed another set of SQGs using the interim sediment quality guideline (ISQG) and the Probable Effect Level (PEL). ISQG represents concentration below which adverse effects on aquatic organisms are expected to be rare, while the PEL corresponds to concentration above which adverse effects are expected to occur frequently (CCME, 1999). Values lying between ISQG and PEL indicate a possible occasional occurrence of biological effects.

For our entire dataset, the concentrations of individual congeners lied well below the ERL and ISQG values of the SQG (*Table 10*). This indicates that the adverse effects exerted by these aromatic hydrocarbons would rarely be observed in organisms.

Table 10: SQG values for ERL and ERM, ISQG and PEL as well as the individual PAH concentration ranges detected in sediment samples in 2023

PAHs	Concentration range (ng g^{-1} dw)			SQG ERL-ERM[a]	% of samples	SQG ISQG-PEL[b]	% of samples
	March	June	September		<ERL		<ISQG
Nap	<LOD – 0.74	<LOD – 5.06	<LOD – 1.12	160 – 2100	100	34.6 – 391	100
Acy	0.04 – 0.76	0.07 – 1.18	<LOD – 0.76	16 – 500	100	5.87 – 128	100
Ace	<LOD – 0.25	<LOD – 0.38	<LOD – 0.31	44 – 640	100	6.71 – 88.9	100
FIu	0.43 – 1.36	0.45 – 2.40	0.14 – 1.66	19 – 540	100	21.2 – 144	100
Phe	2.73 – 10.19	3.40 – 16.17	2.44 – 16.99	240 – 1500	100	41.9 – 515	100
Ant	0.99 – 1.78	0.45 – 1.70	<LOD – 1.96	600 – 5100	100	46.9 – 245	100
Flt	0.70 – 10.74	3.23 – 16.67	3.61 – 21.97	85.3 – 1100	100	111 – 2355	100
Pyr	0.21 – 7.49	2.48 – 12.66	2.66 – 33.69	665 – 2500	100	53 – 875	100
BaA	2.40 – 5.03	1.77 – 5.59	0.77 – 5.56	261 – 1600	100	31.7 – 385	100
Chr	<LOD – 3.69	2.42 – 9.66	0.82 – 6.79	384 – 2800	100	57.1 – 862	100
BbF	3.61 – 9.89	4.09 – 13.93	1.78 – 15.43	320 – 1880	100	-	-
BkF	<LOD – 0.28	0.84 – 3.78	0.35 – 2.74	280 – 1620	100	-	-
BaP	0.38 – 2.31	1.29 – 3.74	<LOD. – 3.20	430 – 1600	100	-	-
DBahA	0.82 – 1.51	1.21 – 2.29	<LOD – 1.64	63.4 – 260	100	6.22 – 135	100
Ind	1.19 – 2.93	1.27 – 5.02	0.39 – 4.39	-	-	-	-

BghiP	1.01 – 4.01	1.22 – 5.59	0.40 – 6.03	430 – 1600	100	-	-

[a] ERL and ERM values were obtained from Bajt (2012), and Mbusnum et al. (2020)

[b] ISQG and PEL values were obtained from CCME (1999)

The second SQG approach is to calculate the TEQ and MEQ relative to the concentration of B(a)P following the formulas previously described. The detailed information on the TEQ and MEQ values for each sampling station can be found in *Annex 30.*

For the samples collected in March, the $\sum TEQ_{C\text{-}PAHs}$ ranged from 1.9 to 5.5 ng g^{-1} dw. The values were not different for the June (3.3 – 9.0 ng g^{-1} dw), and September samples (0.76 – 7.56 ng g^{-1} dw). The $\sum TEQ_{C\text{-}PAHs}$ for Red River sediments were coherent with the values found in Can Gio wetland, Ho Chi Minh City (<LOD – 6 ng g^{-1} dw; Thuy et al., 2018) and an urban river in Hanoi (0.29 – 1.38 ng g^{-1} dw; Toan et al., 2020). The MEQ were in the range of 2.09 – 6.56 ng g^{-1} dw for March, 3.37 – 10.48 ng g^{-1} dw for June, and 1.24 – 9.41 ng g^{-1} dw for September. The stations in the city center or strongly influenced by anthropogenic activities exhibited the highest values of TEQ and MEQ (R05 in March, R04 in June, and R09 in September). The results for TEQ and MEQ in the Red River samples were well below the threshold reported in the literature (Iwegbue et al., 2021; Ugochukwu et al., 2022), implying that the PAHs in the Red River sediment did not pose significant threats.

3.4. Conclusion

This study provides new insights into the occurrence, distribution, and potential risks of PAHs in sediment samples from the Red River across both dry and rainy seasons. Overall, PAH concentrations in the Red River sediment were in low to moderate contamination category, and all values remained well below national sediment quality standards. Moreover, a clear seasonal pattern was observed, with PAH levels generally higher in the rainy season than in the dry season. This suggests that hydrological processes, particularly runoff and enhanced transport during high-flow conditions, play an important role in mobilizing and redistributing PAHs within the river system.

The congener profiles were relatively stable at all sampling periods, being dominated by 4-ring PAHs. LMW PAHs such as Naph et Ace were rarely detected, which is consistent with their high volatility and susceptibility to degradation. Source appointment using

diagnostic ratios and PCA indicated that combustion was the primary source of PAHs, reflecting the influence of biomass burning, vehicle emissions, and fossil fuel combustion associated with both urban, industrial, and agricultural activities along the river.

Risk assessment based on ERL/ERM, ISQG/PEL, and TEQ/MEQ approaches consistently showed that ecological and human health risks posed by PAHs in the Red River sediments are currently very low. This is an important finding, as it demonstrates that despite the increasing pressures from urbanization, industrialization, and extensive agriculture in the Red River Delta, sediment quality in this river has not yet reached levels of high concern.

These findings highlight the need for regular monitoring. This will allow for an early detection of any increases in PAH contamination levels in the river and provide a scientific basis for the development and gradual implementation of future policies. Moreover, further studies that combine chemical monitoring with hydrodynamic and sediment transport modeling are recommended. Such approaches would improve the understanding of how seasonal hydrological regimes influence the transport of PAHs from upstream through the river network and ultimately into the marine environment.

Acknowledgments

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4. Polychlorinated biphenyls, Polybrominated diphenyl ethers, and Chlorinated paraffins.

Context and objective

The previous chapter has established a solid foundation for extending the investigation to other classes of persistent organic pollutants that share similar environmental behaviors. The next chapter shifts the focus from PAHs to PCBs, PBDEs, and CPs, groups of chlorinated and brominated POPs that have been progressively listed and controlled under the Stockholm Convention during the past two decades. Studying these compounds alongside PAHs is crucial, as it allows not only for a comparison between well-characterized and more recently regulated contaminants, but also for a better understanding of how persistence, accumulation, and regulatory measures interact to shape their current distribution and risks in sediments. The chapter 4 thus broadens the perspective on sediment contamination and paves the way for a more comprehensive assessment of contaminant mixtures in the Red River.

Vietnam's ratification of the Stockholm Convention in 2002 marked an important milestone in its efforts to reduce the environmental and health risks posed by POPs. For the last two decades, the country has made significant progress through national inventories of PCBs, PBDEs, and CPs, as well as the implementation of regulatory framework such as nation technical regulation QCVN 43:2017/BTNMT (sediment quality standards for seven indicator PCBs) and decree 82/2022/ND-CP restricting the industrial use of PCBs, deca-BDE, and short-chain CPs. These initiatives show nation's strong commitment to internation obligation in managing POPs.

Nevertheless, the actual occurrence and distribution of these substances in the environment remain insufficiently and sparsely documented. Flowing through industrial

sites, agricultural land, and densely populated areas, the river is highly exposed to potential inputs of hazardous chemicals. Yet, despite its ecological and socio-economic significance , few systematic studies have addressed the levels and profiles of POPs in Red River sediments. The lack of data represents a limitation for evaluating both environmental risks and the effectiveness of ongoing efforts and existing regulations.

This chapter presents an article in preparation that responds to the sub-objective 2 mentioned in section 1.5, aiming at generatin original datasets on PCBs, PBDEs, and CPs, in the Red River sediment, identifying possible contamination sources and examining how these compounds behave in relation to sediment characteristics, anthropogenic pressures, seasonal changes and regulatory. By analyzing three chemical families of industrial use that have had a different marketing and prohibition agenda, the study provides an integrated view of contamination patterns in a river under increasing anthropogenic pressure.

A total of 21 sediment samples were collected along the Red River transect, ensuring spatial representativeness across urban, peri-urban, rural, and estuarine zones. Sediment samples of both seasons (November 2022 and June 2023) were analyzed for PCBs and CPs while PBDEs were analyzed in November samples exclusively.

All sediment samples were subjected to simultaneous extraction using PLE. The extracts were subsequently analyzed by compound-specific gas chromatography techniques coupled with mass spectrometry detection.

Results' overview

The concentrations of PCBs in Red River sediments were consistently low across the two sampling campaigns. In all samples, PCB levels were below 50 ng g^{-1} dw. The PCB concentrations in the samples were well under the Vietnamese national technical regulations for sediment quality, which set thresholds at 277 ng g^{-1} for freshwater sediments and 189 ng g^{-1} for brackish and marine sediments. These results suggest that PCBs do not represent nowadays a major contaminant of concern in the Red River. Their relatively low concentrations show both the historical nature of PCB usage, and their ban and global phase-out following their inclusion as the initial POPs in the Stockholm Convention. The spatial and seasonal variation further indicates that PCBs in the Red River sediments are primarily residues of past contamination rather than evidence of

ongoing emissions. Indeed, PCB levels demonstrated a slight decrease over time, from the dry season (November 2022) to the rainy season (June 2023).

In contrast, CPs were present at considerably higher concentrations, ranging in the hundreds of ng g^{-1} dw. These levels indicate the increasing relevance of CPs as emerging contaminants in the Red River. Unlike PCBs, CPs displayed the evidence of more recent input, with concentrations increasing from dry to rainy season. The temporal increase in levels illustrates ongoing emissions from various sources, likely linked to their widespread applications in industrial materials, notably in polyvinyl chloride (PVC), which has been mass-produced in Vietnam.

Regarding PBDEs, only two out of eight targeted congeners, BDE 47 and BDE 209, were detected in the Red River sediments. PBDEs have been progressively regulated under the Stockholm Convention, with earlier restrictions on penta- and octa-BDE mixtures (2009) and later inclusion of deca-BDE (2017). As a result, their environmental inputs might have declined earlier than those of CPs, which remained in use in Vietnam until 2023. The detection of BDE 209 in only on sample possibly showed its later phase-out (compared with other mixtures) and its susceptibility to degrade into more stable lower-brominated congeners such as BDE 47, which were frequently detected in the Red River samples.

Risk assessment on sediment suggests that the Red River currently exhibits low ecological risks associated with PCBs, CPs, and PBDEs. PCB concentrations were far below sediment quality guidelines and unlikely to cause adverse ecological effects. However, for CPs, moderate risks were observed in some samples collected in June 2023, raising concerns about potential future impacts.

Partial conclusion

This assessment urges greater attention for CPs as a newer class of contamination. While fully regulated PCBs show declining trends, CPs may become the major components in the contamination profile of sediment in Vietnam's aquatic environments. Continued monitoring will therefore be critical to prevent CPs from becoming a significant long-term ecological risk in the Red River system. Overall, contamination levels for these three families of pollutants remain low to moderate compared to other countries. This is consistent with the fact that PAH concentrations determined previously are at the lower

end of the scale. This can be linked to the low organic matter content of the analyzed sediments.

The detailed results and discussions are included in the Publication N°3 under preparation, entitled "Occurrence, Distribution, and Ecological Risk Assessment of Historical and Contemporary Persistent Organic Pollutants in sediments from the Red River (Vietnam)".

Publication N°3: **Occurrence, Distribution, and Ecological Risk assessment of Historical and Contemporary Persistent Organic Pollutants in Sediments from the Red River (Vietnam)**

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**under preparation*

Abstract:

Sediment samples were collected at 21 stations along the downstream 200 km transect of the Red River crossing Hanoi, Vietnam, to investigate the occurrence of halogenated persistent organic pollutants including Polychlorinated biphenyls (PCBs), Polybrominated diphenyl ethers (PBDEs), and Short and medium chain Chlorinated Paraffins (CPs). The average sum concentration of the seven PCBs was in the low range, varying from 7.9 ng g^{-1} dw in November 2022 down to 0.11 ng g^{-1} dw in June 2023. Penta- and Hexachloro biphenyls were the most dominant congeners in both sampling seasons. CPs showed different temporal trend, with higher levels measured during the rainy season (mean: 348 ng g^{-1} dw in June 2023) compared to the dry season (mean 128 ng g^{-1} dw in November 2022), suggesting recent inputs through riverbank leaching. Of the eight PBDEs analyzed, only BDE 47 and BDE 209 were detected in November samples at low levels. The ecological risk assessment, based on sediment quality guidelines, indicated that the overall risk posed by all analyzed compounds is generally low. However, in June, two sites exceeded the threshold for moderate risk, specifically due to the presence of CPs. To our knowledge, this study provides the first baseline assessment of CPs in Vietnamese

river sediments. The contrasting temporal trends between PCB and CPs suggest the recent release of the latter into the environment.

Keywords: Polychlorinated biphenyls, Polybrominated diphenyl ethers, Chlorinated paraffins, spatial-temporal variations, risk assessment.

Highlights:

1. CPs were detected at hundreds ng g^{-1} in the Red River sediments.
2. PCBs show historical contamination patterns, while CPs indicate recent input .
3. Only BDE 47 and BDE 209 were found at detectable levels in the samples.
4. The decline of chemical families in the environment follows their inclusion in the Stockholm Convention.
5. The Red River sediments exhibited low to moderate ecological risks.

4.1. Introduction

A large proportion of contaminants enters river systems directly through soil erosion along the riverbanks, runoff, and atmospheric particle deposition, where pollutants have already been sorbed onto solid matter (Begum et al., 2021; Mohammadi et al., 2018). Once introduced into the water column, hydrophobic organic compounds present in dissolved phase have a strong tendency to partition onto suspended particulate matter, which eventually settles and become incorporated into the sediment, where they can resist degradation for extended period of time (Chiaia-Hernández et al., 2022; He et al., 2014). In this case, sediment acts as a sink, receiving pollutants from several inputs, including soil erosion, surface runoff and untreated urban or industrial discharges (Fuchte et al., 2022), and atmospheric deposition (Castro-Jiménez et al., 2015). Moreover, sediment can release pollutants back into the water column under resuspension or bioturbation events (Roberts, 2012; Banta et al., 2003), depending on the physico-chemical conditions. This dual characteristic, as a sink and a source, makes sediment an essential matrix for the monitoring of the quality status of the aquatic compartment and for assessing the potential long-term risks posed by persistent pollutants.

Vietnam is becoming particularly vulnerable to contamination by anthropogenic organic pollutants. Over the last three decades, the country has undergone rapid economic development and industrial expansion, especially in densely populated regions such as the Red River Delta. The Delta spans about 14300 km2 with a population of 24 million (General Statistics Office of Vietnam, 2024). Alongside the rapid growth of urban-based industries, the Red River Delta has also experienced significant rural industrialization, particularly with the proliferation of craft villages, traditionally specialized in activities such as ceramics, textile dyeing, and food production, has expanded to metallurgy and solid waste recycling (Dang et al., 2020). According to Vietnam's Ministry of Agriculture and Environment in 2025, nearly 30% of craft villages in the country are located in the Red River Delta. Even though industrialization has brought important socio-economic benefits to the country, it also causes major challenges concerning limited capacity of waste treatment and management posing high risk of environmental contamination and accumulation of toxic substances in sediments (World Bank, 2013).

Halogenated organic compounds are among the most concerning groups of industrial chemicals, since they are generally persistent and toxic to humans and living organisms. Due to their resistance to fire, heat, and chemical degradation, compounds such as Polychlorinated biphenyls (PCBs), Polybrominated diphenyl ethers (PBDEs), and Short and Medium-chain chlorinated paraffins (S-MCCPs) have been widely used in the manufacturing industries (Costa et al., 2014; De et al., 2004; McGrath et al., 2023). However, these substances have been proven to exert harmful impacts on human health, including endocrine disruption, immunotoxicity, neurotoxicity, carcinogenicity, and other health issues related to development and reproduction (Buha Djordjevic et al., 2020; Couderc et al., 2016; X. Wang et al., 2019). Their effects, persistence, ability for long-range transport and bio-accumulation, have led to their listing in the Stockholm Convention on Persistent Organic Pollutants (POPs). PCBs are among the first twelve initial POPs to be listed in the Stockholm Convention under Annex A (Elimination). Later on in 2009, commercial penta- and octaBDE mixtures were added in Annex A, followed by the inclusion of deca-BDE in 2017. SCCPs were also listed in 2017 and, more recently, MCCPs were listed in 2025, both in Annex A. Although none of PCBs, PBDEs, and SCCPs have ever been produced in Vietnam, the proximity of the country to China, a major producer of these chemicals, further increases the risk of contamination. Despite the ban on PCB production since 1974, China had produced about 10,000 tons of PCBs, accounting for

0.6% of the global historic production (H. Yu et al., 2023). It was estimated that over 460,000 tons of decaBDE had been produced in China from 1993 to 2018 (J. Wang et al., 2021). In 8 years from 2006 to 2013, China had increased the CP production from 260,000 tons $year^{-1}$ to 1,000,000 tons $year^{-1}$ (Glüge et al., 2016). Moreover, these chemicals are present in Vietnam from a long period of time according to recent inventory efforts reported in the National Implementation Plan submitted to the Stockholm Convention. PCBs had been imported to Vietnam during the second half of the 20th century. Large quantities of PCBs remain in storage facilities. Recent reports indicated that more than 1,300 tons of oil contaminated with PCBs were still present in the country by 2015. For PBDEs, inventories revealed that in 2014, over 568 tons of commercial octa-BDE were estimated in electrical and electronic equipment in Vietnam, while 6.5 tons of commercial penta-BDE mixtures were present in in-use vehicles in 2011. SCCPs were also identified in several hundred thousand tons of plastic in the transport sector as toxic additives (UNEP, 2017). PCBs and PBDEs have been extensively detected in sediments collected in Vietnam (Duong et al., 2014; Giuliani et al., 2019; Hoang, Duong, et al., 2023; Lam et al., 2017; Toan et al., 2020), while CPs were detected in ambient air in Hanoi, the capital city of Vietnam (C. He et al., 2023).

Studying PCBs, PBDEs, and CPs collectively is highly relevant as these compounds share physicochemical characteristics (e.g., persistence, bioaccumulation, and toxicity), were all of industrial use, and subjected to international regulation under the Stockholm Convention, but have had a different marketing and prohibition agenda. As mentioned earlier, although environmental data on PCBs and PBDEs have been reported in Vietnam, information for CPs in aquatic systems is still absent (Vu et al., 2025). The Red River Delta represents an interesting study area as it is among the most densely populated and industrialized regions in Vietnam. The combined impacts of domestic, industrial, agricultural, and domestic activities can cause continuous inputs of pollutants into the aquatic environment. Moreover, the river receives inputs from upstream China, a global PBDEs and CPs production powerhouse, and discharges into the Gulf of Tonkin, representing a potential pathway for freshwater-to-marine pollutant transfer. Given that these persistent organic pollutants are known to undergo long-range transport, reaching remote regions of the Arctic and Antarctica (Eckhardt et al., 2007; Kallenborn et al., 2013), establishing baseline sediment data in the river is essential. This fills the information gap on organic pollutants in Vietnam and enables Vietnamese policy makers to fulfill the

obligations of the Stockholm Convention and develop suitable strategies for pollution control and risk management.

To this end, 21 surface sediment samples of the Red River were collected from the city center of Hanoi to the Ba Lat estuary during dry and wet seasons, to assess the occurrence, congeners' profiles, potential sources, and ecological risks associated with seven PCBs, eight PBDEs, and mixture of CPs in the river sediments.

4.2. Materials and Methods

4.2.1. Study area and sampling campaigns

Originating in China and flowing through northern Vietnam before entering the Gulf of Tonkin by the Ba Lat estuary, the Red River, with a total length of over 1100 km, drains a large catchment having diverse socio-economic activities, such as rice cultivation and agriculture, aquaculture and fisheries, industrial zones, urban centers and rural craft villages (Liao et al., 2024; T. Q. Tran et al., 2021). The Red River Delta is not only one of Vietnam's most densely populated areas (1128 inhab.km^{-2}, 2024), but it also contributes significantly to the agriculture and industrial production (e.g., textiles, construction materials, and electronics) of the nation. Several industrial zones in major cities such as Hanoi, Nam Dinh and Thai Binh are located along the Red River.

Two sampling campaigns were carried out in November 2022 (dry season) and in June 2023 rainy season). Twenty-one sampling sites (named R00 to R20, *Annex 8*) were chosen at intervals of approximately 10 km between the capital city of Hanoi and the Ba Lat estuary, with the total transect of 200 km (*Figure 24*).

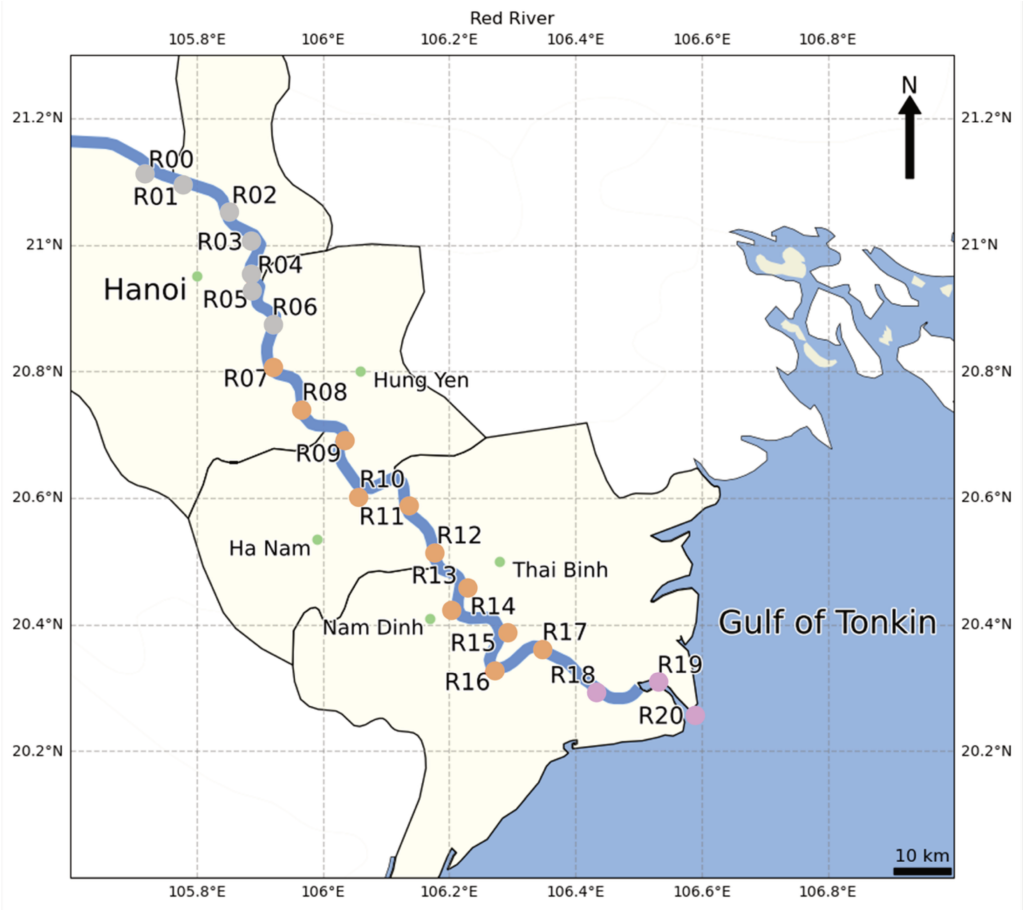

Figure 24 : The Red River and geographical location of the 21 sampling stations from the city center of Hanoi to the Ba Lat estuary (Gulf of Tonkin), divided into three zone (zone 1, upstream Hanoi in grey, zone 2 medium in brown, and zone 3 to the estuary in violet).

The 21 sampling stations can be divided into three geographical zones depending on the anthropogenic activities along the Red River. Zone 1 (Z1) convers the first seven stations (R00 to R06, marked as grey points in *Figure 24*). This zone lies within the center of the capital city of Hanoi (more than 8 million inhabitants) and several industrial zones (especially Thang Long and Bac Thang Long industrial parks specialized in manufacturing of electronics and electrical equipment, vehicles, consumer products and food processing) and craft villages (e.g., Bat Trang village with the production of bricks and ceramic products). Zone 2 (Z2) extends over 11 stations (R07 to R17, marked as brown points in *Figure 24*) and corresponds to the major rural area of the Red River Delta. Smaller cities such as Nam Dinh or Thai Binh (approximately 2 million inhabitants in each city) are nonetheless located in this zone. Zone 3 (Z3) is the shortest section consisting of three stations in the Ba Lat estuary (marked as violet points in *Figure 24*).

Approximately 500 g of surface sediment were collected in the riverbed (3-4 m away from the riverbank) at each sampling stations using a Van Veen Grab Sampler. The samples were stored at -20 ^{o}C in pre-calcinated aluminum containers. The samples were freeze-dried at lab using a Freeze Dryer (Buchi, pro200, Switzerland) and sieved (<200 µm). Afterwards, they were stored in aluminum foil at room temperature prior to extraction and analysis.

4.2.2. Chemical reagents

The standard mixture of the seven PCBs (PCB 28, PCB 52, PCB 101, PCB 118, PCB 138, PCB 153 and PCB 180, mixture PCB-mix 3 at 10 µg mL^{-1} in isooctane) and certified standard for SCCPs (Chloroparaffin C10-C13, 63% of chlorine by weight at 1000 µg mL^{-1}, in cyclohexane) were purchased from LGC Standards. The mixture BDE-CSM in Isooctane/Toluene (80:20, v/v) containing the eight congeners of PBDE (BDE 28, BDE 47, BDE 99, BDE 100, BDE 153, BDE 154, BDE 189 at 20 µg mL^{-1}, and BDE 209 at 200 µg mL^{-1}), in addition to the surrogate standards BDE 79 and BDE 189 and the internal standard BDE 140 (at 50 µg mL^{-1} each, in isooctane) were provided by Accustandard. The surrogate standards d_5-PCB30 and d_3-PCB156 and the internal standard d_5-PCB116 (at 100 µg mL^{-1} each, in isooctane) and the internal standard $^{13}C_6$-hexachlorobenzene (100 µg mL^{-1} in nonane) were from CIL Cluzeau. Dichloromethane (DCM for HPLC, ≥ 99.8%) and hexane (Hex for HPLC, ≥ 97%) were obtained from Sigma Aldrich.

4.2.3. Extraction and Analytical methods

The extraction of PCBs, PBDEs and SCCPs were adapted from Godéré et al. (2022) and Rigal et al. (2024) with slight modifications. The extraction was performed using Pressurized Liquid Extraction (ASE 200 Accelerated Solvent Extractor, Thermo Scientific™ Dionex™, USA) with 33 mL stainless-steel extraction cells. Ten mg of sediment sample was spiked with surrogate standards (20 ng sample^{-1} of d_5-PCB30, d_3-PCB156, BDE 79, and BDE 189). The extraction process used the mixture of Hex/DCM (1:1, v/v) as solvent and underwent two cycles of 7 minutes each at the temperature of 100 and the pressure of 100 bar. The extracts were then concentrated under a gentle air flow until 1 mL and went to the clean-up step on silica gel cartridges (SPE SupelcleanTM 154 LC-Si, 1 g, 6 mL, Supelco). The extract (1 mL) was loaded into the cartridge pre-conditioned with 6 mL

each of DCM, mixture of Hex/DCM (1:1, v/v), and Hex. After that, the cartridge was eluted with 8 mL of Hex for Fraction 1 (F1, containing PCB and BDE) and 8 mL of Hex/DCM (1:1, v/v) for Fraction 2 (F2, containing CPs). Each fraction was thereafter evaporated under a gentle air flow and reconstituted in 200 µL of Hex. Twenty ng sample^{-1} of d_5-PCB 116 and BDE 140 were added to F1 and 20 ng sample^{-1} of 13C6-hexachlorobenzene were added to F2 as internal standards.

PCBs and CPs were analyzed by a gas chromatography (GC-7890A) coupled with tandem mass spectrometry system (MS-3000A, Agilent Technologies) with electron ionization (GC/EI-MS/MS). The analysis of PBDEs was performed using gas chromatography (GC Clarus 600 AutoSystems XL) in combination with electron capture negative ion mass spectrometry (GC/ECNI-MS, MS Clarus 600C, PerkinElmer). The analytical conditions were adapted from the literature and are detailed in *Annex 15*.

4.2.4. Quality assurance and Quality control

All the containers of samples and materials for extraction steps were thoroughly cleaned. The glassware was rinsed with water and acetone and calcinated at 350 ^{o}C for 8 hours. Procedural blanks were performed using Fontainebleau sand in every batch of sample extraction. The mean recoveries of surrogates in sediment samples were 47±11% (d_5-PCB30), 138±34% (d_3-PCB156), 90±11% (BDE 79), and 131±22% (BDE189). The concentrations of detected compounds were adjusted according to the surrogate recovery rate and the blank levels. QC calibration points were injected every 5 samples. The accuracies were within 107±19% for PCBs, 105±12% for PBDEs, and 86±17% for SCCPs. The LOD values evaluated at a signal-to-noise ratio of 3 were from 1.22 to 19.4 pg g^{-1} for PCBs, from 0.004 to 1.01 ng g^{-1} for PBDEs, and 0.86 ng g^{-1} for SCCPs (*Annex 10*).

4.2.5. Risk assessment

The risk quotient (RQ) associated with the presence of PCBs in sediments was calculated as follows:

$$RQ = \frac{MEC}{PNEC}$$

where MEC is the measured environmental concentrations of PCBs in sediments and PNEC is the predicted no-effect concentration. As indicated in the literature (P. C. Le et al., 2022), the PNEC value was the threshold for the sum of 7 indicator PCBs in sediment established in Vietnamese national technical regulation on sediment quality (QCVN-43:2017/BTNMT). The value RQ indicates the potential risk, with value below 0.01 for very low risk, between 0.01 and 0.1 for low risk, between 0.1 and 1 for medium risk and above 1 for high risk. (P. C. Le et al., 2022; Rigal et al., 2024; Toan et al., 2020).

Several Sediment Quality Guidelines (SQGs) have been widely used to assess the ecological risk of organic contaminants (Frignani et al., 2007; Kanzari et al., 2014; Mbusnum et al., 2020; Nouira et al., 2013). In this study, the assessment of PCBs was conducted using SQGs based on effect range low (ERL), effect range medium (ERM), threshold effect level (TEL), and probable effect level (PEL) values. For SCCPs, the Federal environmental sediment quality guidelines (FEsQGs) established by the Canadian government were adopted, since few regulation thresholds are provided.

4.3. Results and discussion

4.3.1. Polychlorinated biphenyls

The sum concentrations of the seven PBCs (∑7PCBs) in sediment samples collected in November 2022 and June 2023 are presented in the *Annex 31*. In November, ∑7PCBs ranged from not detected (n.d.) to 50.5 ng g^{-1} dw with a mean concentration of 7.94 ng g^{-1} dw. In June, concentrations were much lower, ranging from n.d. – 0.37 ng g^{-1} dw, with a mean of 0.11 ng g^{-1}. The PCB levels determined in the Red River were comparable to those in urban rivers in Hanoi (dry season: 0.43 – 13 ng g^{-1} dw, wet season: 0.5 – 13 ng g^{-1} dw; Hoang et al., 2021); (dry season: 2.23 – 8.93, wet season: 0.95 – 4.19 ng g^{-1} dw; Quynh et al., 2019) in An Hoa estuary (1.52 – 19.5 ng g^{-1} dw; Le et al., 2022), and in Saigon River (<LOD – 18.73 ng g^{-1} dw; Babut et al., 2019). Hoang et al. (2021) also suggested the decline of PCB levels in sediment in urban rivers in Hanoi when compared to over one decade ago.

As for November 2022, PCBs were detected in 14 out of the 21 samples. PCB 28, the only trichlorobiphenyl among the targeted compounds, and also the lightest and degradable one (Steliga et al., 2020), was not found in any sample. Overall, pentachloro-, hexachloro-, and heptachlorobiphenyls are more dominant than the tetrachlorobiphenyl

congener PCB 52. Among the seven congeners, PCB 138 was the most abundant, detected in 67% of the samples and accounting for an average of over 43% of ∑7PCBs. It was followed by PCB 180, PCB 153, and PCB 101, which were as often as detected as PCB 138, but contributed at lower proportions (27%, 25%, and 6.7% respectively).

For samples from June 2023, R16 and R19 samples showed no detectable PCB contamination. PCB 28, PCB 52, PCB 101, and PCB 138 were not detected in any sample. PCB 118 exhibited the highest DF at 90.5%, being the predominant congener across nearly all samples. PCB 138 was present in five samples, accounting for an average of over 46% of the ∑7PCBs. PCB 180 was only present in sample R01 where it was the most dominant, contributing more than 43% of ∑7PCB.

The ∑7PCB in sediment samples in November 2022 was significantly different than those in June 2023 (Student's t-test, $p < 0.01$). This indicates that PCB levels were lower in the early rainy season compared to the early dry season. Toan et al. (2020) reported a similar seasonal pattern for sediment samples collected in urban area of Hanoi, revealing historical inputs. Several factors may explain this seasonal difference. During the early dry season, the hydrodynamic conditions of the Red River would be more stable, with reduced flow and less sediment resuspension, which favor the retention of PCBs in sediments. In contrast, the early rainy season is characterized by increased rainfall, surface runoff, and higher discharge, which would enhance solid inputs either airborne or waterborne or from soil, which settled in the riverbed and can mask or dilute historical pollution over short periods of time. Although suspended solids are mobilized by the surface runoff, these particles may not be heavily contaminated with PCBs since PCBs are a group of historical POPs, which were banned globally in the early 2000s, and their use has been highly restricted by the Vietnamese government since 2011 (Decree 26/2011/ND-CP) to comply with the Stockholm Convention. The arrival and the sedimentation of the non-contaminated particles in the riverbed may reduce the PCB concentrations in the whole sediment samples.

Despite the difference in PCB levels, the congener profile remained quite similar between the two sampling periods, with penta- and hexachlorobiphenyls being the dominant congeners found in sediment. Congeners with a high degree of chlorination have strong hydrophobicity which enhances their affinity for organic matter and sediment particles (Llorca et al., 2020; Reddy et al., 2019). As a result, these congeners tend to

persist in sediments at higher proportions compared to lower chlorinated PCBs, which are more water soluble and volatile, and more likely to biodegrade (Beyer et al., 2009; Sakai et al., 2016). The dilution effect during the rainy season and the similarity of congener profiles between both seasons further revealed the historical inputs of PCBs. This PCB profile can be observed in sediments from the coastal areas in Singapore (Wurl & Obbard, 2006), Kim Nguu River in Hanoi (Toan et al., 2020) and An Hoa estuary in central Vietnam (Le et al., 2022).

Pearson's correlation test was performed to investigate the correlation among the PCB congeners detected in the different samples. The detected PCBs in samples collected in November 2022 showed strong positive correlation with each other ($r = 0.75 – 0.99$, $p < 0.01$, *Annex 32*). Forensic experts warned that PCB profiles in sediment are subjected to alteration. Physical processes, e.g., dissolution and volatilization, microbial dechlorination, and weathering (i.e., biodegradation, photolysis, leaching) can progressively modify congener compositions (Erickson, 2020; US-EPA, 2012). Over time, these environmental processes homogenize PCB mixtures and can produce similar profile patterns among samples. Consequently, strong correlations observed in the Red River sediments reflect long-term environmental mixing and homogenization of PCB profiles.

The spatial distribution of the ∑7PCBs according to zone defined along the Red River, according to the map on Figure, is presented in *Figure 25*, for both seasons.

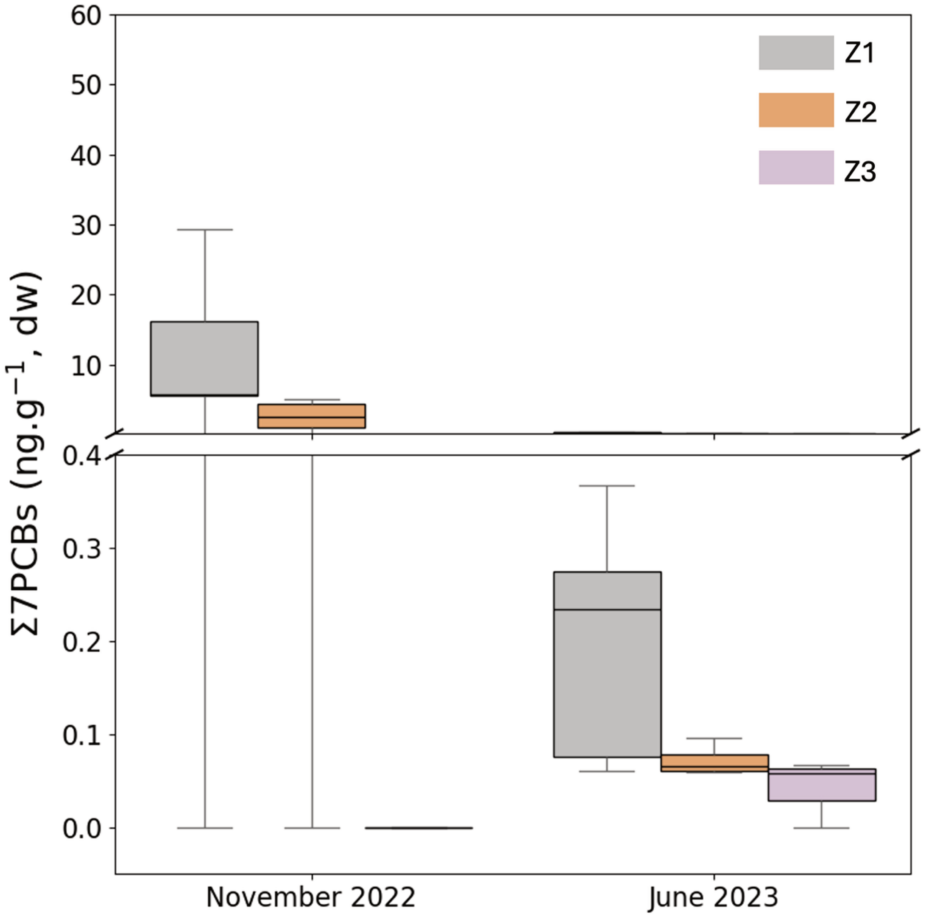

Figure 25 : Box-plots of the sum concentrations of the seven PCBs (∑7PCBs) measured in sediments during the dry (November 2022) and wet (June 2023) seasons at the 21 stations, grouped according to three geographical zones: Z1 (urban center of Hanoi), Z2 (sub-urban and rural zones of the Red River Delta), and Z3 (Ba Lat estuary).

Overall, PCB concentrations showed a clear decreasing trend from the urban center of Hanoi towards the Gulf of Tonkin for both sampling periods. In November 2022, average PCB concentrations decreased by approximately 28% from Z1 to Z2, eventually reaching non-detectable levels at the Ba Lat estuary. A similar downstream decline was observed in June 2023 samples, with a sharp reduction of about 78.5% between Z1 and Z3. The spatial pattern in sediments suggests that Hanoi and upstream can have contributed to the initial input of PCBs to the Red River. Moreover, the possible presence of PCB-containing equipment storage and special disposal facilities in the surroundings of Hanoi may justify the higher levels detected in Z1. Further downstream in the rural area, the dilution effect and limited additional inputs could contribute to the decrease in PCB concentrations. These findings are in agreement with previously published spatial

fingerprints. In major French rivers, PCB concentrations typically peaked in metropolitan or industrial centers and progressively decreased toward the estuarine zones. For example, PCB hotspots have been identified along the Seine River between Paris and Rouen, followed by a sharp decrease in levels near the estuary at Le Havre (Dendievel et al., 2020). A similar pattern was seen in the Rhone River, where elevated PCB levels were measured in sediments collected near Lyon, with a gradual decrease further downstream in the lower Rhone section (Dendievel et al., 2020).

The ∑7PCBs is a parameter that can be used to assess the quality of the aquatic compartments and the environmental risk. Based on the sum PCB concentrations in sediment, the quality status can be classified into five levels: Background (Level I) with ∑7PCBs < 5 ng g^{-1} dw; Good (Level II), 5 – 17 ng g^{-1} dw; Moderate (Level III), 17 – 190 ng g^{-1} dw; Poor (Level IV), 190 – 1900 ng g^{-1} dw; and Very Poor (Level V), > 1900 ng g^{-1} dw (Jartun et al., 2010; Kanzari et al., 2014). According to this classification, for November 2022, three samples exhibited a moderate level of quality, six a good level, and five were at background level. For June 2023, all samples fell within the background level.

The assessment of potential environmental risks posed by PCBs in sediment in the Red River was conducted based on RQ and ERM/PEL. The results are presented in *Table 11* and *Table 12*.

Table 11 : RQ values of PCBs in the Red River sediments

	MEC (ng g^{-1} dw)	PEC (ng g^{-1} dw)	RQ	% of samples		
				RQ < 0.1	0.1 ≤ RQ < 1	RQ > 1
November 2022	n.d. – 50.5	277*	n.d. – 0.18	90.48	9.52	-
June 2023	n.d. – 0.37		n.d. – 0.0013	100	-	-

*threshold for the sum concentrations of the seven PCBs in freshwater sediment set in national technical regulation on sediment quality (QCVN-43:2017/BTNMT)

Table 12: SQG values for PCBs in the Red River sediments.

				% of samples				% of samples	
	∑7PCBs	ERL*	ERM*	under ERL	ERL to ERM	TEL*	PEL*	under TEL	TEL to PEL
November 2022	n.d. – 50.5	22.7	180	90.52	9.48	21.5	189	90.52	9.48
June 2023	n.d. – 0.37			100	-			100	-

*Values obtained from Le et al. (2022)

The RQ linked to PCBs in the November sediment samples ranged from n.d. to 0.18. Detailed data on RQ can be found in *Annex 33*. Two samples (R00 and R12) fell into the medium-risk category, while the remaining November samples as well as all June samples were scored at low risk. The ERL/TEL guideline values lead to similar results, suggesting that ecotoxicological effects, associated to PCB contamination in sediments of the Red River, are unlikely to occur.

4.3.2. Short-chain chlorinated paraffins

The total concentrations of SCCPs (∑SCCPs) detected in the Red River sediment samples are reported in *Annex 34* and their spatial-seasonal variations are shown in *Figure 26.* The ∑SCCPs in November 2022 samples ranged from <LOD to 293.6 ng g^{-1} dw with a mean of 127.6 ng g^{-1} dw. SCCPs were detected at 15/21 sampling stations, since at R13, R15 and R17-R20 stations, levels were below the detection limit. In June 2023, ∑SCCPs were detected at 19/21 stations, at higher concentrations, from <LOD to 1515 ng g^{-1} dw with a mean of 348.0 ng g^{-1} dw, with only two stations (R12 and R19) below the LOD. To the best of our knowledge, these data of SCCPs concentrations in sediment samples are the very first ones reported whether in Vietnam or in the Southeast Asian region, whereas SCCPs were determined in ambient air in Hanoi (<0.39 – 44 ng m^{-3}, C. He et al., 2023) and in indoor dust from Thailand (median value of 29 µg g^{-1}; McGrath et al., 2023). The SCCP levels in the Red River sediments were comparable to those in rivers passing through major cities of Tokyo (100.4 – 248.9 ng g^{-1} dw), Osaka and Kyoto (4 – 161.3 ng g^{-1} dw) in Japan (Iino et al., 2005), in rivers (n.d. – 2020 ng g^{-1} dw) in industrial city of Shanghai (X. T. Wang et al., 2019) but much lower than those detected in Xiaoqing River in China (mean value: 1300 ng g^{-1} dw). However, ∑SCCPs in our study were higher

than those in the Ravi river (<LOD – 170 ng g^{-1} dw) in Pakistan (Tahir et al., 2024) or in major rivers (27.4 – 146 ng g^{-1} dw) in South Korea (Choo et al., 2020).

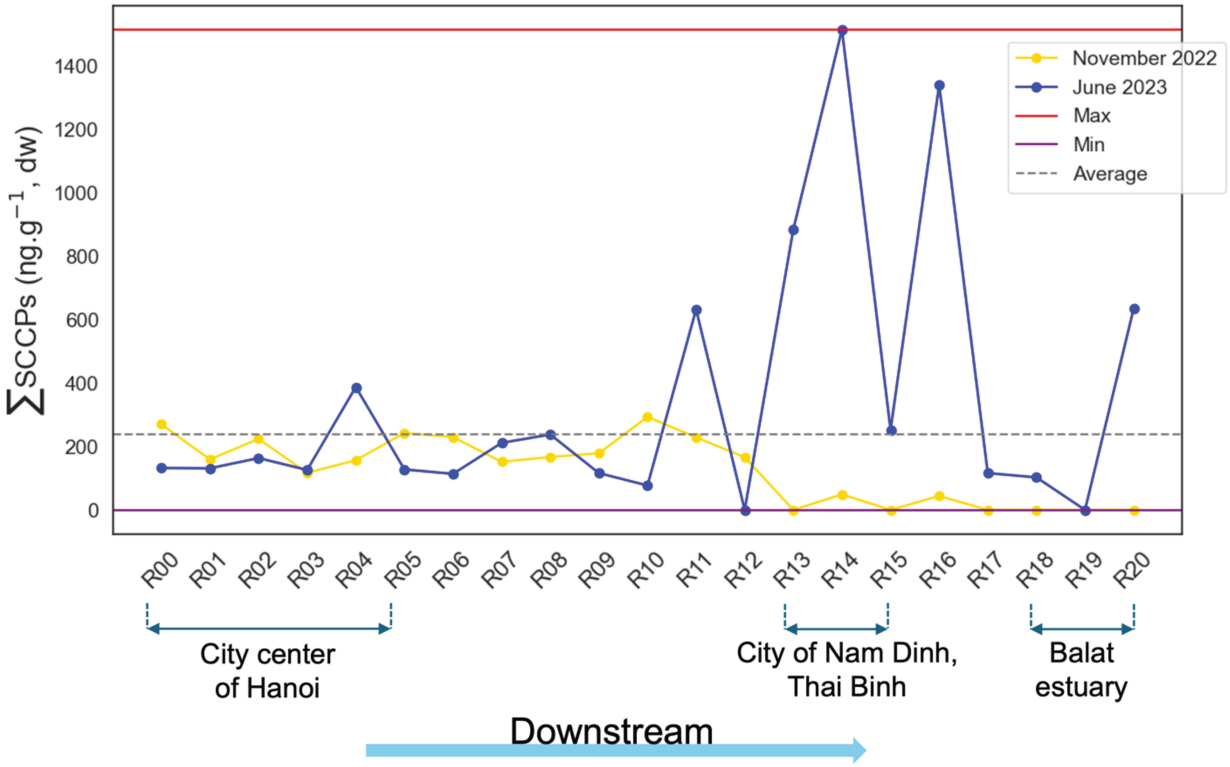

Figure 26 : Concentrations of SCCPs (∑SCCPs) determined in the Red River sediments during the dry (November 2022, in yellow) and the rainy (June 2023, in blue) seasons.

The ∑SCCPs measured in June were significantly higher than in November ($p < 0.05$). This clear seasonal difference suggests inputs of SCCPs during the early rainy season, where stronger hydrological activity likely mobilizes contaminants from terrestrial sources into the riverbed. Interestingly, this trend of SCCPs is the opposite of that observed for PCBs at the exact sampling locations, indicating that SCCPs reflect the ongoing emission rather than historical contamination.

SCCPs were only listed in 2017 as POPs in the Stockholm Convention, and the restrictions in Vietnam are more recent, with regulations in the industrial sectors coming into force in 2022. Additionally, Vietnam registered for an exemption under the Stockholm Convention for the continued use of 15000 tons of SCCPs per year until December 2023 (https://www.pops.int/Implementation/Exemptions/SpecificExemptions/ShortchainchlorinatedparaffinsRoSE/tabid/7595/Default.aspx, accessed August 2025). This very recent regulatory implementation can be an explanation for the ongoing release of SCCPs

in the environment. Unlike PCBs, which have been strictly regulated decades ago, SCCPs were still in use until very recently and are still present in many consumer products.

Actually, SCCPs are primarily applied as plasticizers in PVC (Guida et al., 2020), one of the most widely produced plastics in Vietnam. In 2018, the national demand for PVC was estimated at over 2.2 million tons, accounting for 29% of the total plastic consumption (Ta et al., 2021). It was estimated that nearly 2 million tons of PVC had been for the manufacturing of building materials, electronics and electrical equipment, and in civil construction in 2018. Moreover, it has been shown that SCCPs occur at high proportions in PVC and rubber products (Tran et al., 2025; Vetter et al., 2023). Thus, Vietnam's role as a major PVC producer and consumer strongly suggests a present release of SCCPs into the environment. Furthermore, Nguyen et al. (2024) reported that PVC accounted for 25.8% and 14.2% of microplastics detected in water and sediment samples from the Red River, respectively. These findings provide clear evidence that PVC debris are a dominant contributor to the plastic pollution in this river. Given that SCCPs are embedded in the plastics, leaching and weathering of PVC and e-waste are likely to gradually release these chlorinated contaminants into the river system. Hydrological conditions during the rainy season may accelerate this release by increasing particle transport and plastic fragmentation (Liro et al., 2023), which could explain the higher concentrations of SCCPs measured in sediment samples from June.

The spatial distribution of SCCPs in sediments along the Red River shows clear differences between the two sampling campaigns and among the river zones (from Z1 to Z3, *Figure 27*).

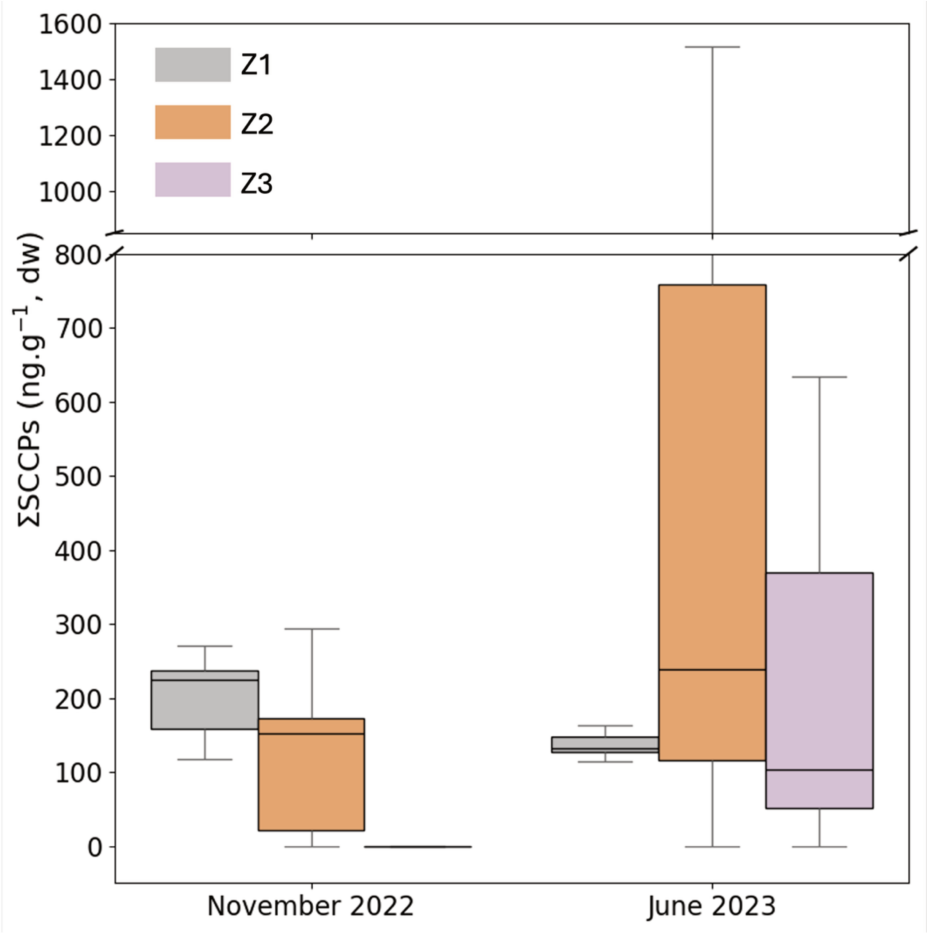

Figure 27: Box-plots of the sum concentrations of SCCPs (∑SCCPs) measured in sediments during the dry (November 2022) and wet (June 2023) seasons at the 21 stations, grouped according to three geographical zones

In November 2022, a decreasing trend downstream towards the ocean can be observed as shown in (*Figure 27*). The highest levels of SCCPs were consistently detected in samples collected both in the urban center and peri-urban areas of Hanoi, whereas concentrations declined markedly in Z2 and reached values below the detection limit in Z3. On average, ∑SCCPs decreased by 41% from Z1 to Z2, before dropping to non-detectable levels in the estuarine zone. This pattern is similar to the one's registered for PCBs in both sampling periods, suggesting that during the early dry season, contamination of SCCPs was primarily localized in urban and industrial hotspots within Hanoi. On the contrary, in June, a different spatial pattern was observed. In Z1, concentrations remained relatively stable compared to November, with no significant temporal differences ($p > 0.05$). However, in Z2, the average level of SCCPs nearly tripled relative to Z1, and the maximum level exceeded 1500 ng g^{-1} dw. The highest concentrations were detected at stations R13 and

R14, located near Nam Dinh and Thai Binh cities. Although Z2 is generally considered a rural zone, these results pointed to localized industrial activities, notably the textile industry in Nam Dinh, representing potential active sources of SCCPs (Tomasko et al., 2023). From Z2 to Z3, SCCP levels decreased by 50%, though there was a slight peak at station R20 (635 ng g^{-1} dw), suggesting additional downstream inputs. Overall, November was characterized by an urban-centered contamination gradient, while June revealed stronger contamination levels downstream, with Z2 as the most impacted zone. The spatial distribution of SCCP in Z2 is consistent with previous observations of microplastic contamination in the Red River, where high abundances of microplastics have been reported in both water and sediment in this zone (T. T. Nguyen et al., 2024). This once again highlights the strong association between plastic materials and SCCPs, which are used as plastic additives. The absence of seasonal variation in Z1 suggests relatively constant inputs in the Hanoi metropolitan area. The sharp increase in Z2 during June showed more seasonal-specific and localized release of SCCPs.

To assess the ecological risk posed by the presence of SCCPs in the Red River sediments, we relied on the Federal Environmental Quality Guidelines for chlorinated alkanes proposed by Environment Canada (2016). These guidelines classify SCCPs concentrations as low risk (< 1800 ng g^{-1}), moderate risk (1800 – 18,000 ng g^{-1}), and high risk (> 18,000 ng g^{-1}), values being normalized to 1% organic carbon. ∑SCCPs normalized to 1% of the total organic carbon are given in SI for the Red River samples (*Annex 8* and *Annex 34*) and results are plotted on the *Figure 28*. Nearly all sediment samples in November fell under the low-risk classification. However, in June, two sites (R14 and R16) exceed the 1800 ng g^{-1} threshold, indicating a moderate ecological risk. Although the overall ecological risk caused by SCCPs in Red River sediments may currently be considered as low, our data suggesting actual release and the presence of sites ranked at moderate-risk in June 2023 highlight a concerning issue. With the continued use of CPs in manufacturing processes in Vietnam and their actual release into the environment, the upcoming escalation of ecological risks posed by chlorinated paraffins in the Red River is highly probable.

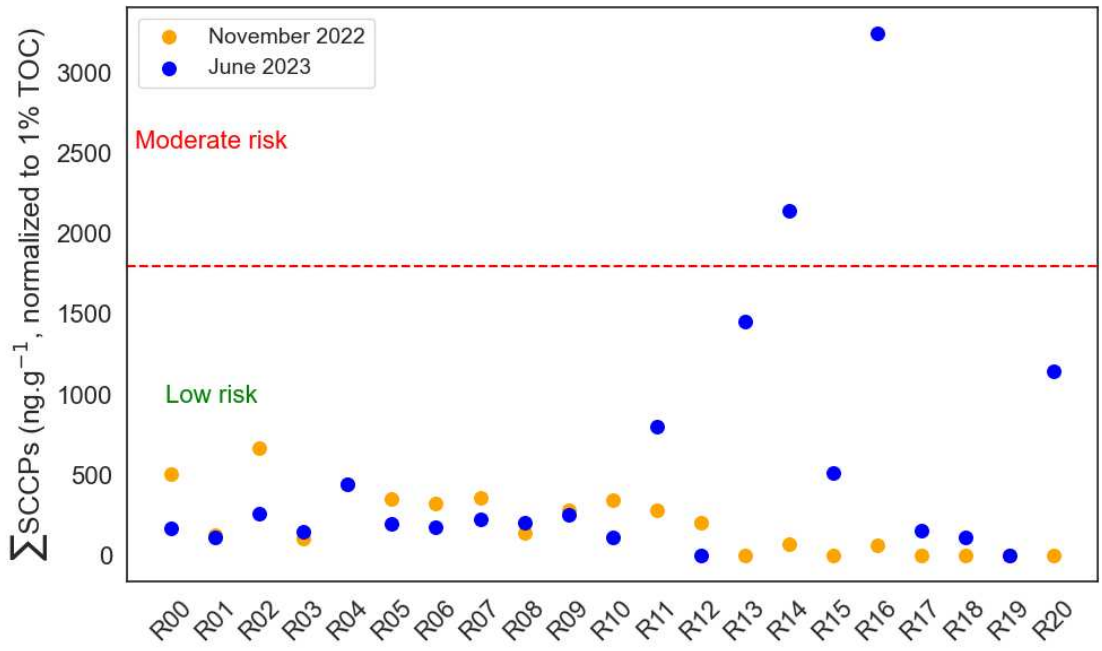

Figure 28 : Sum concentrations of SCCPs normalized to 1% TOC (∑SCCPs, normalized to 1% TOC) measured in the sediments during the dry (November 2022, yellow) and wet (June 2023, blue) seasons at the 21 stations along the Red River. The dotted red line at 1800 ng g^{-1} is the threshold for low to moderate risk according to the Canadian Environmental sediment quality guidelines (Environment Canada, 2016).

Besides SCCPs, medium-chain chlorinated paraffins (MCCPs) have also been used as plasticizers, as extreme pressure additives in metal working fluids and as additives to polymeric materials (Glüge et al., 2018; UNEP, 2023a). They have been investigated in every continents (Glüge et al., 2018) and are increasingly recognized as contaminants of emerging concern. Several studies have reported MCCPs in the atmosphere, water, sediment, soil, and living organisms, highlighting their wide distribution and persistence in the environment (Labadie et al., 2019; McGrath et al., 2023; van Mourik et al., 2016; Wei et al., 2016). Given their similar physicochemical properties and potential for long-range transport, MCCPs exhibit similar behavior in the environment as SCCPs (Guida et al., 2020; X. T. Wang et al., 2019). Therefore, the Stockholm Convention has adopted decision SC-12/10 in May 2025, listing MCCPs in the Annex A (Elimination) together with SCCPs (UNEP, 2025). This milestone marks the global regulation of MCCPs production and use. As a consequence, it is critical to incorporate both SCCPs and MCCPs into routine monitoring programs. The analytical method used in this study is also capable of detecting and measuring MCCPs together with SCCPs (Godéré et al., 2022). However, for a more

comprehensive understanding of sources, profiles, and risks, congener-specific determination remains more accurate and informative. Such an approach will facilitate comprehensive assessments of chlorinated paraffins pollution in the Red River, and more broadly across the Vietnamese environment, providing profound scientific evidence driving future regulatory actions in the country.

4.3.3. Polybrominated diphenyl ethers

The total concentrations of the eight targeted PBDEs (∑8PBDE) were presented in *Table 13.* ∑8PBDE in sediment samples collected in November 2022 ranged from n.d. to 7.33 ng g^{-1} dw. Among the eight congeners, BDE 47 was the most abundant and could be detected in all samples (DF = 100%) at the concentration between 0.017 – 0.047 ng g^{-1} dw. It was followed by BDE 100 (DF = 23.8%), BDE 28 and BDE 209, whose concentrations were higher than LOD in only one sample for each congener (R06 and R03, respectively). One notable exception was sample R03 where BDE 209 (7.27 ng g^{-1} dw) dominated the PBDE profile accounting for more than 99% of ∑8PBDE. The dominance of BDE 209 in this sample could imply the localized and temporal input of deca-BDE contamination.

BDE 47 is the major component of the commercial penta-BDE technical mixture, historically used in polyurethane foams, mattresses and upholstered furniture (Costa et al., 2014). However, BDE 99, which is normally present at higher proportions than BDE 47 in this formulation, assessed at level below LOD in all of the samples. A similar pattern was reported in Monastir Bay in Tunisa where BDE 47 was the only detectable PBDE and BDE 99 was absent (Nouira et al., 2013). The authors explained this observation by the allochthonous inputs of BDE 47, possibly originating from long-range atmospheric transport and deposition (Mariani et al., 2008; Ruth Ohoro et al., 2021). In addition, BDE 47 may also be formed through the debromination of higher brominated congeners (Ilyas et al., 2011; Sánchez-Prado et al., 2005), particularly BDE 99 and BDE 209, under photolytic or microbial degradation processes, which further contributes to its prevalence in sediments

Table 13: PBDEs concentrations (ng g^{-1} dw) in sediment samples collected in November 2022 and June 2023

Samples	BDE 28	BDE 47	BDE 99	BDE 100	BDE 153	BDE 154	BDE 183	BDE 209	∑8PBDE
R00	<LOD	0.034	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.034
R01	<LOD	0.024	<LOD	<LOD	n.d.	<LOD	<LOD	<LOD	0.024
R02	<LOD	0.017	<LOD	<LOD	n.d.	n.d.	<LOD	<LOD	0.017
R03	<LOD	0.047	<LOD	0.016	n.d.	<LOD	<LOD	7.27	7.331
R04	n.d.	0.021	<LOD	n.d.	n.d.	<LOD	n.d.	<LOD	0.021
R05	<LOD	0.025	<LOD	<LOD	n.d.	<LOD	<LOD	<LOD	0.025
R06	0.020	0.028	<LOD	<LOD	n.d.	n.d.	<LOD	<LOD	0.048
R07	<LOD	0.018	<LOD	<LOD	n.d.	<LOD	<LOD	<LOD	0.018
R08	<LOD	0.043	<LOD	0.016	<LOD	<LOD	<LOD	<LOD	0.059
R09	<LOD	0.026	<LOD	0.014	<LOD	<LOD	<LOD	<LOD	0.039
R10	<LOD	0.025	<LOD	<LOD	n.d.	n.d.	<LOD	<LOD	0.025
R11	<LOD	0.038	<LOD	0.016	<LOD	<LOD	<LOD	<LOD	0.054
R12	<LOD	0.034	<LOD	0.018	<LOD	<LOD	<LOD	<LOD	0.051
R13	n.d.	0.018	<LOD	<LOD	n.d.	n.d.	<LOD	<LOD	0.018
R14	n.d.	0.024	<LOD	<LOD	n.d.	n.d.	<LOD	<LOD	0.024
R15	n.d.	0.028	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.028
R16	n.d.	0.017	<LOD	<LOD	n.d.	<LOD	<LOD	<LOD	0.017
R17	n.d.	0.019	<LOD	<LOD	n.d.	n.d.	<LOD	<LOD	0.019
R18	n.d.	0.022	<LOD	<LOD	n.d.	<LOD	<LOD	<LOD	0.022
R19	n.d.	0.018	<LOD	<LOD	n.d.	<LOD	<LOD	<LOD	0.018
R20	n.d.	0.024	<LOD	<LOD	n.d.	n.d.	<LOD	<LOD	0.024

The results of the present study align with those reported in one sediment sample also collected in the Red River by Hoang et al. (2021). Only BDE 47 and BDE 209 were detected in this sample, with concentrations in the dry season at 0.0032 and 0.2 ng g^{-1} dw, for congeners 47 and 209, respectively. The PBDEs levels reported by the same authors for urban rivers flowing in city center of Hanoi were much higher than for the Red River, with

the ∑8PBDE reaching up to over 55 ng g^{-1}. BDE 209 was the major congener, accounting for more than 90% of the total concentrations. Babut et al. (2019) analyzed PBDEs (BDE 209 was not included) in sediments from the Saigon River in Ho Chi Minh city, another metropolis in Vietnam, but the levels were all below the LOD.

PBDEs were progressively regulated under the Stockholm Convention, with penta- and decaBDE mixtures listed in 2009 and decaBDE mixture, which predominantly consists of BDE 209, only added later in 2017. Accordingly, their environmental inputs have been declining earlier than those of CPs (especially SCCPs which had been used in Vietnam until 2023). The fact that BDE 209 was found in only one sample may reflect the later phase-out of decaBDE use and the environmental behavior of BDE 209. Although in use until recently, BDE 209 is prone to photolytic or microbial debromination, yielding more stable lower-brominated congeners such as BDE 47 (Akortia et al., 2016; S. Fang et al., 2025). Importantly, PBDEs and CPs share strong associations to plastic materials. PBDEs were used as flame retardant additives in plastics, textiles, and other materials. Commercial penta-BDE mixtures were primarily incorporated into polyurenthane foams, typically at concentrations of 3-5% by weight (Jinhui et al., 2017). Octa-BDE mixtures were used mainly in acrylonitrile-butadiene-styrene polymers, usually at 12-18% by weight (Jinhui et al., 2017). DecaBDE mixtures were predominantly used in electrical and equipment, such as computer, television sets and other household appliances (Jinhui et al., 2017). As mentioned above, SCCPs are mainly incorporated as secondary plasticizers in PVC, rubber, sealants, and coatings. Despite the similar applications, the regulatory timeline could help explain the contrasting sediment profiles of PBDEs and SCCPs. PBDEs were listed under the Stockholm Convention between 2009 and 2017, resulting in a gradual decline of environmental inputs. Their current presence in Red River sediments therefore likely reflects residual contamination from legacy uses or debromination of BDE-209. In contrast, SCCPs remained in use in Vietnam until 2023, and thus continue to be released from plastic materials and other applications, leading to their broader and more recent presence in the sediment compartment.

4.4. Conclusion

To the best of our knowledge, this is the first investigation of the simultaneous presence of these three groups of POPs in the sediments from the downstream section of

the Red River. The study provides a comprehensive assessment of PCBs, PBDEs and CPs in the Red River sediment, offering new insights into the occurrence, spatio-seasonal variations, and ecological risks of these halogenated compounds used in industry over the past four decades. PCBs, which are historical POPs put under regulations for long time, exhibited very low concentrations (n.d. – 50.5 ng g^{-1} dw) and spatio-seasonal trend reflecting pollution with higher levels during dry season, which acknowledge the effectiveness of regulatory efforts worldwide and in Vietnam. PBDEs, which were introduced more recently but were regulated between 2008 (penta- and octaBDE mixtures) and 2017 (decaBDE mixture), were detected at low levels (n.d. – 7.3 ng g^{-1} dw), with only BDE 47 and BDE 209 found at detectable levels in dry season sediments exclusively. On the other hands, SCCPs were detected at higher concentrations (<LOD – 1515 ng g^{-1} dw), with seasonal trend showing significant increase during the rainy season in comparison to the dry season. This implies on-going release of CPs into the environment from both point and non-point sources in the surroundings of the Red River. Based on the contents of PCB and SCCPs and the corresponding SQGs, most of the samples from the two sampling periods presented low ecological risk. However, some sampling sites in June show moderate ecological risks posed by SCCPs. This emerging risk is concerning given the upward temporal trend of SCCPs and MCCPs, which may result in more elevated ecological threats in the near future, since emissions are expected to persist.

The difference in trends between PCBs and SCCPs gives a bigger picture of hazardous chemical management in Vietnam: historical legacy pollutants, such as PCB or organochlorinated pesticides, are declining under regulatory efforts, whereas more emerging groups, such as SCCPs and chlorinated paraffins (CPs) in general, remain insufficiently tracked. Vietnam has, up to now, established clear regulatory frameworks for PCBs but it is not the case yet for PBDEs and CPs, particularly SCCPs. The establishment of more comprehensive monitoring programs, alongside the implementation of clear environmental quality guidelines, would greatly support Vietnam in addressing POP contamination. Such measurement results could enhance the country's ability to monitor pollution incidents, inform risk assessments, and ultimately achieve the objectives set by the Stockholm Convention.

5. Per- and Polyfluoroalkyl Substances

Context and objective

Following the work on PCBs, PBDEs, and CPs, the chapter 5 takes into account PFAS, a class of highly persistent organic pollutants of growing global concern. Per- and Polyfluoroalkyl substances (PFAS) are organic pollutants recently recognized for their environmental persistence, bioaccumulation potential, and rapidly growing global concern. While a few compounds (PFOA, PFOS, and PFHxS) are listed under the Stockholm Convention, a wide range of PFAS and associated substances remain under evaluation, with limited documentation in Vietnam.

Investigating these substances in the Red River allows for the assessment of both currently regulated PFAS and emerging alternatives, further emphasizing the evolving landscape of chemical contamination in the Red River.

Moving on from the assessment of chlorinated and brominated POPs in chapter 4, chapter 5 studies the presence and distribution of 53 PFAS in both water and sediment samples collected along the downstream section of the Red River. By integrating regulated substances, emerging alternatives, and precursor compounds, chapter 5 aims to achieve the sub-objective 3 in section 1.5 for PFAS of the doctoral and contributes an additional and essential new piece to the picture of organic contamination status in the Red River. Four sampling campaigns were carried out between November 2022 and September 2023. Samples were analyzed using a combination of the target approach and total oxidizable precursors assay (TOPA) to capture not only known PFAS but also previously unquantified precursors.

Methodological approach complementary to target analysis

TOPA is a sample digestion method where polyfluorinated PFAS or perfluoroalkyl acids (PFAA) precursors are oxidized in the presence of hydroxyl radicals ($OH^{\bullet}$). The $OH^{\bullet}$ is generated by the thermal activation of persulfate $S_2O_8^{2-}$ under the alkaline conditions. The generation of $OH^{\bullet}$ is as follows:

$$S_2O_8{}^{2-} \xrightarrow{t^o,\ pH>12} 2SO_4{}^{-\bullet}$$

$$SO_4{}^{-\bullet} + OH^- \rightarrow SO_4{}^{2-} + OH^\bullet$$

The generated OH• will then react with oxidizable precursors to form PFAA as described in *Figure 29.*

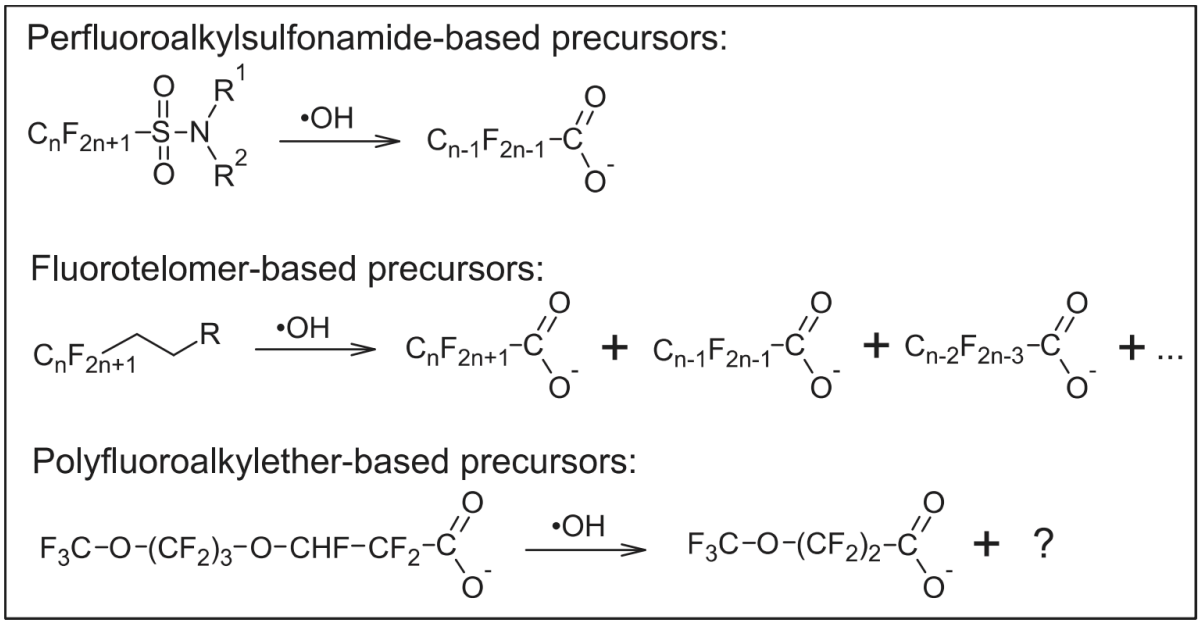

Figure 29: The reaction of PFAA precursors during TOPA (figure obtained from Lange et al., 2024).

The samples (both from target and TOPA approaches) were analyzed for 53 PFAS by liquid chromatography coupled with high-resolution mass spectrometry (Thermo Ficher ScientificTM, Vanquish Flex, OrbitrapTM Exploris 120.

Results' overview

In sediment samples, PFBA and 6:2-FTS were the only detected PFAS, each in one sample from June and September 2023, respectively. On the other hand, all analyzed water samples were contaminated with PFAS, with a maximum reported concentration reaching up to 572 ng L^{-1}. Target analysis revealed the presence of a variety of PFAS, including well known and regulated perfluoroalkyl acids (PFAAs) such as PFOS, PFOA, and PFHxS, as well as emerging polyfluorinated alternatives like 6:2 fluorotelomer sulfonic acid (6:2-FTS) and 6:2 fluorotelomer phosphate diester (6:2-diPAP). The detection of these compounds shows shifting manufacturing and regulatory practices, with the phase-out of PFAS under the Stockholm Convention leading to the introduction of replacement substances that are

now entering the environment. This trend further indicates the global challenge of "regrettable substitution" where regulated hazardous chemicals are replaced with structurally related alternatives that may pose similar risks but remain insufficiently understood.

Beyond the insights provided by target analysis, the application of TOPA revealed the hidden contribution of oxidizable PFAS precursors to the overall contamination profile. Following oxidation, an increase in PFAA concentration was consistently observed in both water and sediment samples. Fluorine equivalent before TOPA could only represent a small fraction of that after oxidation, especially for sediment samples. The TOPA results concluded that there had been a presence of unknown precursors in the environmental samples. For instance, the magnitude of PFHxA formation in several samples highlighted the significant role of 6:2-FTS and related fluorotelomer compounds as precursor sources in the Red River.

The detailed results and discussion are presented in the publication N°4 under preparation, entitled "Poly- and Perfluoroalkyl substances: A First Glimpse of the "Forever Chemicals" in Water and Sediment Samples from Red River, Vietnam".

Partial conclusion

Chapter 5 establishes baseline data for PFAS in the Red River and demonstrates the added value of integrating TOPA with conventional target approaches. By providing new evidence on both legacy and emerging PFAS, as well as their precursors, the findings of chapter 5 reinforce the contamination landscape description in the Red River developed throughout this thesis, including both regulated and historical contaminants (PAHs, PCBs, and PBDEs) and emerging groups (CPs and PFAS). The PhD thesis shows the coexistence of declining contaminants under regulatory efforts and the growing relevance of insufficiently regulated emerging compounds. Overall, the results presented in the review article and across chapters 3 to 5 provide complementary perspectives on the status of organic contamination in the Red River and address past pollution legacies and the current threats. The chapter 5 hence closes the series of case studies on different groups of compounds and opens the conclusion section where major results, perspectives on methodological and analytical aspects, and implications for environmental monitoring and chemical management in Vietnam will be discussed.

Publication N°4: **Poly- and Perfluoroalkyl substances: A First Glimpse of the "Forever Chemicals" in Water and Sediment Samples from Red River, Vietnam**

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**under preparation*

Abstract:

Per- and Polyfluoroalkyl substances (PFAS) are highly persistent organic pollutants of growing global concern, yet their presence remains largely under-documented in Vietnam. This study provides an assessment of 53 PFAS in water and sediment from the downstream Red River. Samples were collected during four campaigns between November 2022 and September 2023 and analyzed by employing the target approach and total oxidizable precursors assay (TOPA). PFAS were detected in water in the range 0.2 - 572 ng L^{-1} while perfluorobutanoic acid (PFBA) and 6:2-FTS were the only two PFAS detected in sediments, each one time in one sample. After TOPA, a clear formation of perfluoroalkyl acids (PFAAs) was observed in both matrices, with perfluoroalkyl carboxylic acids (C_5 – C_{10}) consistently appearing as newly formed PFAS. The notable increase in fluorine equivalents (F_{eq}) after oxidation revealed considerable contributions

from previously undetected PFAS precursors. In several samples, the presence of perfluorohexanoic acid (PFHxA) might indicate the presence of 6:2-FTS and related fluorotelomer compounds. The difference of F_{eq} before and after oxidation highlights an underestimation in PFAS burden in environmental samples using target analysis and requires the integration of non-target approach to better evaluate PFAS contamination.

Keywords: PFAS, TOPA, precursors, water, sediment, Red River

Highlights:

- 53 PFAS were analyzed in Red River water and sediment samples.
- PFAS were detected in every water samples but rarely detected in sediments.
- Alternative PFAS were found alongside regulated ones.
- TOPA revealed the hidden burden of untargeted PFAS precursors.

5.1. Introduction

Poly- and Perfluoroalkyl substances (PFAS) are synthetic fluorinated organic compounds that contain at least one fully fluorinated methyl or methylene carbon atom (without any H/Cl/Br/I atom attached to it, OCED, 2021). Since their development in the 1940s, PFAS have been widely used in industrial and commercial applications, including water-repellent and waterproof materials, lubricants, food packaging, non-stick cookware, textiles, and furniture (Dickman & Aga, 2022; Glüge et al., 2020; Herzke et al., 2012). Additionally, PFAS are key ingredients in Aqueous Film-Forming Foams, which are frequently utilized in airports, military bases, and fire training facilities (Ateia et al., 2023; Houtz et al., 2016). Their continuous use in such settings has driven extensive and globalized PFAS contamination of the surrounding soil, groundwater, and surface waters (Barzen-Hanson 2017). Studies have reported their presence across diverse matrices, such as surface waters, drinking and bottled waters, soils, sediment, and even food (Evich et al., 2022; K. Singh et al., 2023b). Their widespread dissemination has raised significant public health concerns, as humans can be exposed through the consumption of contaminated water and food. PFAS exposure has been associated with wide spectrum of adverse outcomes, including cancers, disruptions to endocrine, immune, neurological,

and reproductive systems, and other health disorders (Blake et al., 2020; Fenton et al., 2021).

Growing evidence of their persistence, bioaccumulation, and harmful health effects has led to the global strategies for the control on the PFAS use. Under the Stockholm Convention on Persistent Organic Pollutants (POPs), perfluorooctane sulfonic acid (PFOS) and its derivatives were the first to be listed in Annex B (Restriction) in 2009, allowing only limited uses in metal plating and fire-fighting foam for liquid fuel vapor suppression and liquid fuel firs (decision SC-9/4); and aiming at reducing emissions. A decade later in 2019, perfluorootanoic acid (PFOA), its salts, and related substances were added to Annex A (Elimination). Most recently, in 2022, perfluorohexane sulfonic acid (PFHxS) and its related compounds were also included in Annex A, highlighting the ongoing global effort to phase out high-risk PFAS. Long-chain perfluoroalkyl carboxylic acids (PFCAs, C_9 – C_{10}) are currently being evaluated for potential inclusion under the Stockholm Convention.

The increasing recognition of PFAS as POPs has led to regulatory actions in several countries. The Drinking Water Directive 2020/2184 of the European Union has set a limit for the total concentration of 20 PFAS, including 10 PFCAs and 10 perfluoroalkyl sulfonic acids (PFSAs) at 100 ng L^{-1}. In the United Sta tes, the Environmental Protection Agency's National Primary Drinking Water Regulation in 2024 set much stricter limits, with PFOA and PFOS each regulated at 4 ng L^{-1} and PFHxS at 10 ng L^{-1} in drinking water (US-EPA, 2024a). Meanwhile, China, a major PFAS producer, has also begun to introduce regulatory standards, setting limits for PFOA and PFOS in drinking water at 80 ng L^{-1} and 40 ng L^{-1}, respectively (J. Zhang et al., 2025). Southeast Asian countries, on the other hand, are only beginning to take steps toward managing PFAS. Current efforts have largely focused on developing national inventories to track the use and importation of PFAS whereas clear regulations for PFAS concentrations in environmental matrices and drinking water are still lacking (Vu et al., 2025).

Vietnam, as a member state of the Stockholm Convention, has shown its commitment to the international obligations. Efforts have been made to inventory the presence of PFOS in the country. According to the National Implementation Plan submitted to the Stockholm Convention in 2017, PFOS has never been produced in Vietnam but imported in the country. It was estimated between 1998 and 2013, PFAS usage in Vietnam ranged from 0.11 to 3.45 tons per year in the textile and upholstery industries, 0.2 to 4.9 tons per year

in paper industry, and 10 to 15 tons per year in firefighting foam in Vietnam (UNEP, 2017). Consequently, PFOS and other PFAS have been detected in various environmental matrices in Vietnam, such as water (Duong et al., 2015; J. W. Kim et al., 2013; Tröger et al., 2021), sediment (Babut et al., 2019; Lam et al., 2017; H. Le Tran et al., 2022) and biota (Hoa et al., 2022; Murakami et al., 2011; Vi et al., 2022). Moreover, many previous studies focused on the region of the Red River Delta (Duong et al., 2015; J. W. Kim et al., 2013; Lam et al., 2017; Phung et al., 2021). The Red River Delta, with a total area of 14300 km^2 and a population of 24 million (in 2024 according to Vietnam's General Statistics Office), is one of the most densely populated regions in Vietnam. The Delta takes its name from the Red River. Originated in the mountainous region of Yunnan province in China, the Red River (total length of 1200 km) flows through the northern Vietnam and discharges in the Gulf of Tonkin. The Red River passes through Hanoi, the capital city of Vietnam (8.7 million inhabitants), and other important provinces in the Delta such as Hung Yen (1.3 million inhabitants), Thai Binh (1.8 million inhabitants), and Nam Dinh (1.8 million inhabitants) (data reported in 2024 by Vietnam's National Statistics Office, https://www.nso.gov.vn/en/population/, accessed September 2025). Urban areas, craft villages, and industrial zones situated along the Red River, together with Gia Lam Airport, represent important potential sources of PFAS. Airports are particularly recognized as hotspots due to the historical and ongoing use of aqueous film-forming foams (AFFFs) in firefighting activities (Filipovic et al., 2015; Houtz et al., 2016), while industrial factories and craft villages may release PFAS through textile dyeing, metal plating, plastics and e-waste recycling processes (Guardian et al., 2020; Phung et al., 2021). Furthermore, the insufficient efficiency of wastewater treatment plants and sewage systems in the region (Hoang, Takahashi, Da Le, Duong, et al., 2021) highlights the risk of PFAS entering the aquatic environment. Hence, systematic monitoring of the Red River is not only critical for understanding the extent of PFAS contamination but also for supporting the development of pollutant management and mitigation strategies.

However, most research and studies focus on limited target list of well-known PFAS namely PFCAs, PFSAs, perfluoroalkyl sulfonamides and fluorotelomer sulfonic acids, which represents only a small fraction of more than eight thousand PFAS of identified structures (Evich, 2022). Moreover, the United States Environmental Protection Agency (US-EPA) has listed more than 14,000 chemicals in its PFAS list, making it practically impossible to analyze all compounds due to the limited availability of analytical standards

and labeled isotopes (https://comptox.epa.gov/dashboard/chemical-lists/pfasstruct, accessed August 2025).

The Total Oxidizable Precursor Assay (TOPA, Houtz et al., 2012) aims to address this limitation by converting oxidizable PFAS precursors into Perfluoroalkyl acids (PFAAs), which are usually included in standard target lists. More importantly, TOPA uses the same analytical instruments and methods as those used in the target approach, making it highly accessible to laboratories and enabling its application in diverse matrices. Initially, TOPA was used to evaluate urban runoff waters, but its application has expanded to a diversity of environmental and biological matrices, including surface water, groundwater, wastewaters, soils, sediment, aquatic biota, and even human fluids (Cioni et al., 2022; Göckener et al., 2021b; Houtz et al., 2013, 2016; Martin et al., 2019). By comparing concentrations measured in target and TOPA approach, the contribution of oxidizable and untargeted PFAS can be evaluated (Göckener et al., 2022b; Lange et al., 2024). An increase in the total PFAA concentration after oxidation indicate the presence of precursors in samples. This approach captures not only precursors that have already been recognized but it also gives the preliminary information of those remaining unidentified or poorly understood.

Given the limited information on PFAS contamination in water and sediment in the water resources in Vietnam and the vulnerability of the Red River Delta, we applied both targeted analysis and TOPA to assess the occurrence of PFAS in water and sediment samples from the Red River, between Hanoi and the Ba Lat estuary. Studying sediment samples allows the assessment of the chronic contamination and long-term accumulation of PFAS while the analysis of water samples provides insights into PFAS fugacity and mobility. This study allows the quantification of well-known PFAS and the evaluation of the contribution of oxidizable precursors to the overall PFAS burden in the riverine samples.

5.2. Materials and Methods

5.2.1. Sampling campaigns

Four sampling campaigns were conducted in November 2022, March, June and September 2023 along the downstream transect of the Red River, spanning approximately 200 km from Hanoi to the Ba Lat estuary in the Gulf of Tonkin. Sediment

was collected in 13 sites in November and 11 sites in the other three campaigns. In parallel, water samples were taken during selected campaigns and chosen for analysis. The map of sampling stations in the Red River is presented in *Figure 30*.

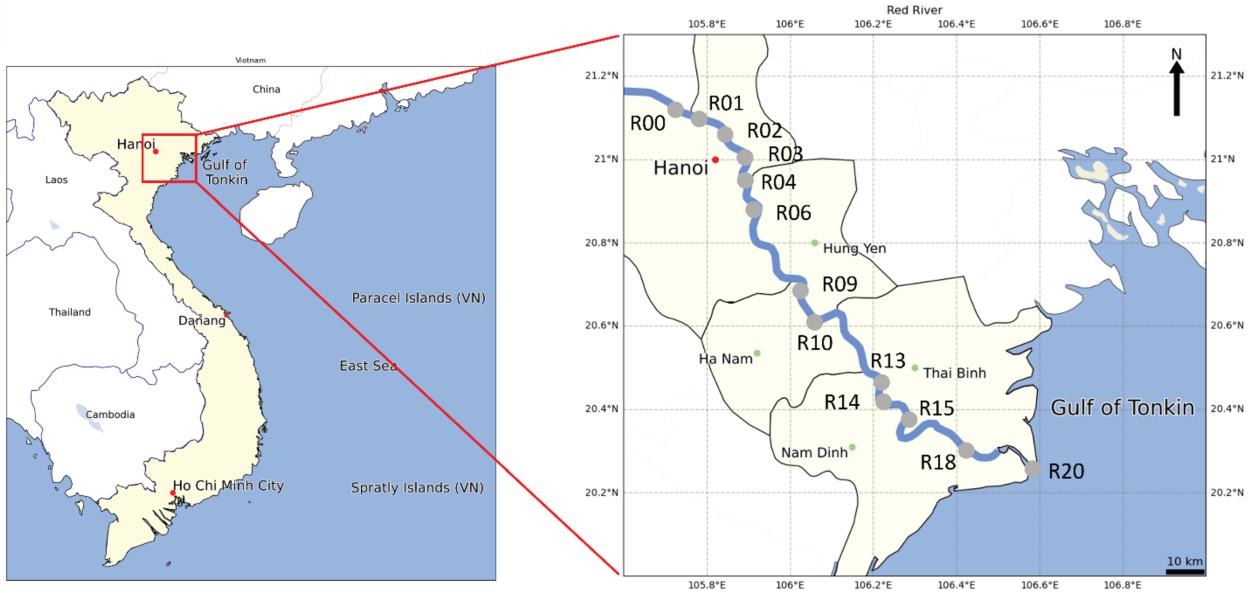

Figure 30: The Red River and the sampling stations (Red River Delta, Northern Vietnam).

5.2.2. Chemicals and reagents

The standard solutions of 53 native PFAS (*Annex 11*), surrogate standards (extraction mixture of 24 labelled PFAS, named s-PFAS, MPFAC-HIF-ES), and internal standards (injection mixture of 7 labelled PFAS, named i-PFAS, MPFAC-HIF-IS) were purchased from Wellington Laboratories. Methanol (MeOH, Optima™ LC/MS grade) was obtained from Fischer Chemical. Hydrochloric acid (HCl, 37%) was from Merck KGaA. Ammonium hydroxide (NH_4OH, 25%), Sodium hydroxide (NaOH), sodium persulfate ($Na_2S_2O_8$), and Superclean™ ENVI-Carb™ were purchased from Sigma Aldrich. Fontainebleau sand, 15 mL and 50 mL polypropylene (PP) tubes were from VWR chemicals. 2 mL glass vials and PP lids were from Fischer Scientific.

5.2.3. Extraction methods

5.2.3.1. Sediment samples

The sediment was sampled using a Van Veen grab. The upper 5 cm sediment in the riverbed were collected and put into pre-combusted aluminum boxes (500 °C for 8 hours). The samples were stored in the freezer (-80 °C) and later freeze-dried for 5 days

using a Buchi Lyovapor L-200. Then the dried samples were sieved to a size class inferior to 200 µm using a stainless-steel mesh and stored in pre-combusted aluminum boxes prior to further extraction and analysis. For both targeted and TOPA approaches, a total of 46 sediment samples (*Annex 35*) were analyzed: 13 in November 2022, and 11 (the same as prior, except R10 and R13) from each of the campaigns conducted in March, June, and September 2023.

5.2.3.1.1. Target approach

Sediment samples (1 g dw) were spiked with surrogate standards (s-PFAS at 2.5 – 50 ng sample^{-1}, *Annex 12*). The samples are then extracted with 5 mL of 0.3% NH_4OH in MeOH by ultrasonication for 30 min. After centrifugation at 3500 rpm for 10 min, the supernatant was transferred to a 50 mL PP tube. The extraction step was repeated two more times, each lasted for 10 min. The combined extract was then treated with 10 mg of activated carbon (ENVI Carb), mixed for 5 min, and centrifuged at 3500 rpm for 10 min. The supernatant was transferred and diluted to 250 mL with UPW. After being pH-adjusted, the sample was passed through the Oasis WAX (Waters™, 150 mg, 6 cc, particle size: 30 µm) cartridges pre-conditioned with 4 mL fractions of 1% ammonium hydroxide (NH_4OH) in methanol (MeOH), MeOH and ultrapure water (UPW). After the sample loading, the cartridges were washed with 4 mL UPW and vacuum-dried for 5 minutes. Next, the cartridges were eluted using 5 mL of 1% NH_4OH in MeOH. The extracts were then evaporated under gentle flow of N_2 gas until a volume of 500 µL. After the addition of internal standards (i-PFAS at 2.5 – 10 ng sample^{-1}, *Annex 13*), the extracts were filtered using nylon filter (17 mm diameter, 0.2 µm pore size) and transferred into injection vials.

5.2.3.1.2. Total oxidizable precursors assay (TOPA)

TOPA approach for sediment samples were adapted from Göckener et al. (2022a) and Houtz et al. (2013). Sediment samples (100 mg, dw), 4.76 g of $Na_2S_2O_8$ (200 mM) and 2 g of NaOH (500 mM) and 100 mL of UPW were added into the glass bottle and incubated at 85 °C overnight. The samples were then collected and centrifuged at 3500 rpm for 10 min and later diluted with 100 mL UPW to obtain the final volume of 200 mL. After being pH-adjusted (pH between 5 and 9) and spiked with the surrogate standards (solution MPFAC-HIF-ES at 2.5 – 50 ng sample^{-1}, *Annex 12*), the SPE protocol described in section 5.2.3.1.1 was applied.

5.2.3.2. Water samples

Water samples were collected using a stainless-steel collector and transferred into pre-combusted (500 °C, 8 hours) 1-L glass bottles. The samples were filtered by glass fiber filter (GF/F, 0.7 µm, Whatman™) and stored in freezer at -80 °C before extraction. A total of 23 water samples (*Annex 35*) were chosen to be analyzed: 6 from November 2022, 9 from March 2023, and 8 from June 2023.

5.2.3.2.1. Target approach

Surrogate standards (solution MPFAC-HIF-ES at 2.5 – 50 ng sample^{-1}, *Annex 13*) were added to the samples. Water samples (250 mL) were passed through Oasis WAX cartridges after preconditioning and subsequent elution as described in section 5.2.3.1.1.

5.2.3.2.2. TOPA approach

The TOPA procedure was adapted from (Houtz & Sedlak, 2012). 250 mL of filtered water sample were transferred into a glass bottle. In each bottle, 4 g of $Na_2S_2O_8$ and 3.8 mL of NaOH 10 M were added as oxidizing agents. The bottles were placed in the oven at 85 °C overnight to promote oxidation. Afterwards, the samples were cooled down to room temperature, and the pH was then adjusted between 5-9 with HCl solution (37%). The samples were subsequently subjected to solid phase extraction using Oasis WAX cartridges following the protocol described in Section 5.2.3.1.1.

5.2.4. Analysis

The analysis of PFAS was performed by reverse phase liquid chromatography coupled with high-resolution mass spectrometry (Thermo Ficher Scientific™, Vanquish Flex, Orbitrap™ Exploris 120). The separation was achieved on DIONEX Acclaim™ C_{18} (2.1 mm x 150 mm x 3 µm) column at 40 °C. The injection volume was 10 µL and the total running time was 19 min. The mobile phase was 2mM of ammonium acetate + 0.1% acetic acid in H2O (phase A) and 2mM of ammonium acetate + 0.1% acetic acid in MeOH (phase B). Elution was performed under gradient conditions A/B: 0 min 95:5, 0.5 min 95:5, 3 min 50:50, 11 min 0:100, 16 min 0:100, 16.1 min 95:5. The flow rate was kept constant at 0.4 mL min^{-1}. Mass parameters are detailed in *Annex 16*.

5.2.5. Calculation for the Fluorine equivalent

To better evaluate the contribution of targeted oxidizable precursors and the potential presence of non-targeted ones, the concentrations of PFAS detected in the target approach and those of PFAS detected post TOPA were converted to Fluorine equivalent (F_{eq}, ng F L^{-1} for water and ng F g^{-1} dw for sediment) for each quantified PFAS and eventually summed up for each sample. The conversion is calculated using the equation as follows:

$$F\ equivalent\ (F_{eq}) = \frac{C_{PFAS}\ .\ n_F\ .u_F}{M_{PFAS}}$$

where C_{PFAS} is the individual concentration of detected PFAS in water (ng L^{-1}) or in sediment (ng g^{-1} dw), n_F is the number of fluorine atoms present in the PFAS chemical structure, u_F is the atomic mass of Fluorine (the value of 19 was used in this study), and M_{PFAS} is the molecular mass of PFAS.

5.2.6. Quality assurance and Quality control

All the containers for sample storage and extraction were cleaned and rinsed with UPW and methanol. Procedural blanks (UPW and Fontainebleau sand) were performed together with real samples during extraction processes. For TOPA approach, UPW and Fontainebleau sand were passed through oxidation similar to the real samples. Spiked water and sediment samples using native and surrogate compounds were performed to obtain the recovery rates of the methods (both target and TOPA). The recovery rates are presented in *Annex 11*. The real samples were spiked systematically with s-PFAS as mentioned above, which were later quantified using the i-PFAS. The recovery rates of surrogates were in range of 42±14% to 113±27% (detailed in *Annex 12*) for 46 samples of sediment and 23 samples of water analyzed. The results were corrected according to the recovery rates of surrogates and blank levels. Calibration points were added every 10-sample injection during an analysis batch quality control. The accuracies were between 89±18% and 118±17%. The LOD values were determined based on the European guidelines (Wenzl et al., 2016) and presented in *Annex 11*.

5.3. Results and discussion

5.3.1. Occurrence of targeted PFAS in the Red River samples

5.3.1.1. Sediment samples

PFAS in sediments were generally below the detection limit. Two exceptions were recorded: sample R00 from June 2023 which contained PFBA at 0.66 ng g^{-1} dw, and sample R04 with 6:2-FTS at 1.47 ng g^{-1} dw. It must be taken into account that the method used in this work based on the consensus method EPA-1633 that covers PFAS with a large range of K_{ow}. Long-chain PFAS ($\geq C_9$) were included in the target list but not detected in the sediment samples. To the best of our knowledge, Lam et al. (2017) conducted the first research working on the presence of PFAS in sediment samples in Vietnam. The authors reported that only 3/10 sediments samples from rivers across Vietnam contains PFAS above the LOD, with the concentrations of 8 detected PFAS (4 PFCAs: C_{10} – C_{13}, and 4 PFSAs: C_4, C_6, C_8, and C_{10}) in range of n.d. – 1.86 ng g^{-1} dw. On the other hand, all samples collected from canals in wastewater treatment plants and landfills were contaminated with PFAS, mostly long-chain PFCAs (C_{10} – C_{13}) and PFSAs, 0.18 – 23.4 ng g^{-1}, Babut et al. (2019) detected 14 PFAS in sediments from the Saigon River (<LOD – 1.18 ng g^{-1} dw) where long-chain PFCAs (C_{11}, C_{13}, and C_{14}) and polyfluorinated compounds (e.g., FOSAA, N-EtFOSAA, and 6:2-diPAP) were the most dominant.

5.3.1.2. Water samples

The concentrations of PFAS in surface water samples collected from the Red River are shown in *Figure 31* and the complete data is shown in *Annex 36*, *Annex 37*, and *Annex 38*. The total PFAS concentration in water ($\sum 53PFAS_{water}$) ranged from 0.4 to 572 ng L^{-1} (average concentration = 136 ng L^{-1}) for samples collected in November 2022, from 0.2 to 11.0 ng L^{-1} (average concentration = 2.0 ng L^{-1}) for March 2023, and from 0.2 to 5.3 ng L^{-1} (average concentration = 1.9 ng L^{-1}) for June 2023.

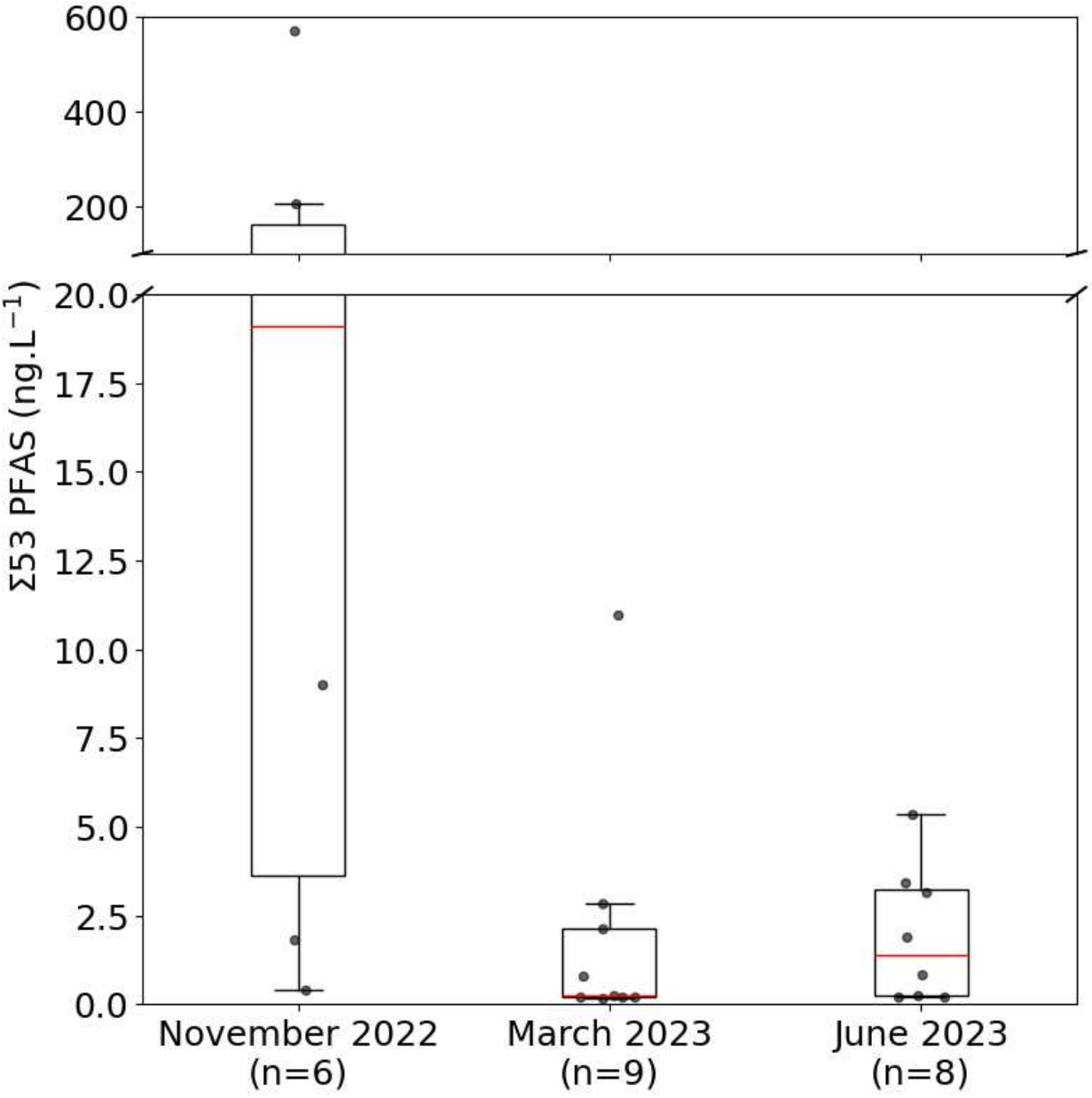

Figure 31: Sum concentrations of the targeted PFAS (∑53PFAS) in water samples from three sampling campaigns.

At least one PFAS was detected in all samples. Out of the 53 targeted PFAS, eight were detected in water samples collected in November 2022. Among PFAAs, PFHxA, PFHpA, and PFOA were identified. Although PFOA was detected in 100% of the samples, it contributed less than 21% to the $\sum 53PFAS_{water}$ in all samples, except for sample R01, where only 3 PFAS were quantified. In addition to PFAAs, several emerging PFAS—particularly fluorotelomers—were also detected in the analysis. The concentration of 6:2-FTS (detection frequency, DF = 83%) could reach up to 203 ng L^{-1} in station R06 and 557

ng L^{-1} in station R14. Six PFAS were detected in March 2023 (5 PFCAs and 1 PFSA). PFOA (DF = 100%) was the most abundant compound, followed by PFHpA (DF = 89%). Though the short-chain PFAS i.e., PFBA and PFPeA were each found in only one sample (R04), they contributed significantly to $\sum 53PFAS_{water}$, accounting for 69 and 9%, respectively. PFBS was the only perfluorinated sulfonic acid detected. It dominated, however, the PFAS profile at stations R06 and R09, contributing up to 92% and 89% of $\sum 53PFAS_{water,}$ respectively. As of samples collected in June 2023, ten PFAS were found. Consistent with the previous sampling campaigns, PFOA and PFHpA were the most frequently detected PFAAs. HPFO-DA (Gen-X) was also among the most predominant PFAS, with a DF of 62.5% and contributing an average of 26% to $\sum 53PFAS_{water}$. It was followed by PFBA (DF = 37.5%) and 6:2-diPAP (DF = 37.5), which represented 31.5% and 21% of $\sum 53PFAS_{water}$ in R01, R02, and R03, respectively.

Kim et al. (2013) were the first to investigate the presence of 17 PFAS (16 PFAAs and FOSA) in surface water in Hanoi. The total levels of 13 detected PFAS could reach up to 170 ng L^{-1} in one sample collected close to an e-waste recycling site. Duong et al. (2015) detected four PFAAs out of a total of 16 targeted PFAS in 14 water samples also collected in the Red River in March 2013. The $\sum PFAS_{water}$ was in range of 0.18 to 1.23 ng L^{-1} with the average concentration of 0.48 ng L^{-1}, where PFOA was the most dominant PFAS. This pattern is consistent with the samples collected in March 2023 in the present study, almost exactly ten years later. Moreover, the PFAS levels measured in this study (0.2 – 571.5 ng L^{-1}) were of the same order of magnitude as those previously reported in highly impacted environments around Hanoi, including urban canals and rivers near dumping sites (2.1 – 360 ng L^{-1}), landfills (103 – 328 ng L^{-1}), and industrial areas (0.37 – 59.5 ng L^{-1},Kim et al., 2013; Ngoc et al., 2021; Phung et al., 2021). This suggests that PFAS levels in the Red River basin may have increased over the past decade (Duong et al., 2015; J. W. Kim et al., 2013), probably reflecting the combined effects of rapid industrial development, urban expansion, and rising consumption of PFAS-containing products in the region.

The PFAS levels in November 2022 water samples (early dry season) show no significant difference with the levels recorded in samples collected in the late dry season (March 2023) and early rainy season (June 2023) (Welch's t-test, $p > 0.05$, *Annex 39, Annex 40,* and *Annex 41*). There were notable differences in the PFAS composition across the three sampling periods. In November 2022 samples, 6:2 FTS was present and contributed

to more than 90% of $\sum 53PFAS_{water}$ in four samples. Several emerging PFAS were also detected in samples from June 2023, contributing to more than 71% of $\sum 53PFAS_{water}$ in R01. Nevertheless, the PFAAs predominated $\sum 53PFAS_{water}$ in March 2023. The temporal variations in PFAS composition likely reflect the occasional releases of PFAS at specific locations along the Red River. This is especially evident from the pronounced spike of 6:2-FTS concentrations detected in November 2022, suggesting a localized or time-specific source of pollution.

Previous studies on PFAS in Vietnam have primarily focused on PFAAs. PFOA and PFOS, the two most extensively monitored PFAS in environmental matrices (Wee et al., 2023), were consistently reported as the dominant compounds in water samples from Vietnam (Duong et al., 2015; J. W. Kim et al., 2013; Lam et al., 2017). In addition to the widespread occurrence of PFOA and PFOS, Lam et al. (2017) also identified the presence of several long-chain PFAS, such as PFDA, PFUdA, and PFTrDA, in certain water samples across the country. However, PFOS was not detected in any of the water samples analyzed in this study. Interestingly, the present study showed a presence of emerging PFAS in the water, especially 6:2-FTS and 6:2-diPAP. It is noteworthy that 6:2- FTS and 6:2-diPAP have been used as emerging alternatives for regulated PFAS, i.e., PFOA, PFOS, and PFHxS which have been subject to global phase-out efforts according to the Stockholm Convention since 2009. These substitutes are commonly found in consumer products (e.g., food-contact materials, papers, and cosmetics) replacing PFOA and PFOS in many industrial application (Brunn et al., 2023; D'eon et al., 2009). The detection of emerging PFAS in water samples indicates a recent shift in industrial practices in the region or more specifically a transition toward the use of alternative PFAS compounds. This trend is likely driven by the regulatory measures implemented by the Vietnamese government, notably the restrictions imposed in 2022 on the production and trading of PFOA and PFHxS in the industrial sector.

Overall, no obvious trend of $\sum 53PFAS_{water}$ could be seen between the upstream and downstream stations along the Red River from all the three period. However, samples from the three sampling campaigns showed similar spatial patterns. Higher PFAS concentrations were detected at monitoring stations located near major cities and residential areas along the river. For example, $\sum 53PFAS_{water}$ in station R06 and R14 in November 2022 reached to 572 ng L^{-1} and 206 ng L^{-1}, respectively. Station R06 is situated near several residential zones on the outskirts of Hanoi. Station R14 is located in the

proximity to Nam Dinh city, another major city in the Red River Delta famous for its textile industry. Moreover, this station serves as the discharge point of the Kien Giang River, which is connected to Thai Binh City's canals and flows through its residential neighborhoods. For samples in March 2023, the highest PFAS level was observed in station R04. This station is situated adjacent to the discharge outlet of Yen So wastewater treatment plant, which is one of the largest facilities in Hanoi. In the samples collected in June 2023, elevated PFAS concentrations were detected in R01, R02, and R03 stations, all of which are located within the central urban area of the capital city. The consistent spatial pattern observed across the three sampling campaigns strongly suggests that urban centers and associated anthropogenic activities play a significant role in contributing to PFAS contamination in the region. This specific trend was also observed in urban rivers and lakes in Beijing (Y. Wang et al., 2019), the Seine River in Paris (Munoz et al., 2018) or other urban zones in France (Munoz et al., 2015). To further investigate the PFAS profiles along the Red River, the principal component analysis (PCA) was conducted and PCA biplot was presented in *Figure 32*. The factor loadings of each PFAS to the two principal components were presented in the *Annex 42*. The first component (PC1) explained for 39% of the variance while the second component (PC2) accounted for 21%. PC1 is strongly associated with PFCAs (e.g., PFHxA, PFHpA, and PFOA) and FTS. These PFAS are highly associated with the industrial applications and usually present in the fire-fighting foams (Herzke et al., 2012; Munoz et al., 2017). R04 (March 2023) and R14 (November 2022) are the two samples with the highest positive scores on PC1. This suggests that these locations are influenced by the discharge of industrial sources or firefighting activities. The compounds driving PC2 with positive loadings are short-chain PFAAs (PFBA, PFPeA) and polyfluorinated compounds, namely, FOSAA, N-EtFOSAA, 6:2-diPAP, and 8:2-diPAP. These compounds are typically used in consumer products such as food packaging or carpets (D'eon et al., 2009; Hoang, Tran, et al., 2023). The prominence of these precursors in urban June samples (R01 to R03, and R06), which corresponds to the early rainy season, implies that increased precipitation and runoff from urban areas may be their primary transport pathway to the river.

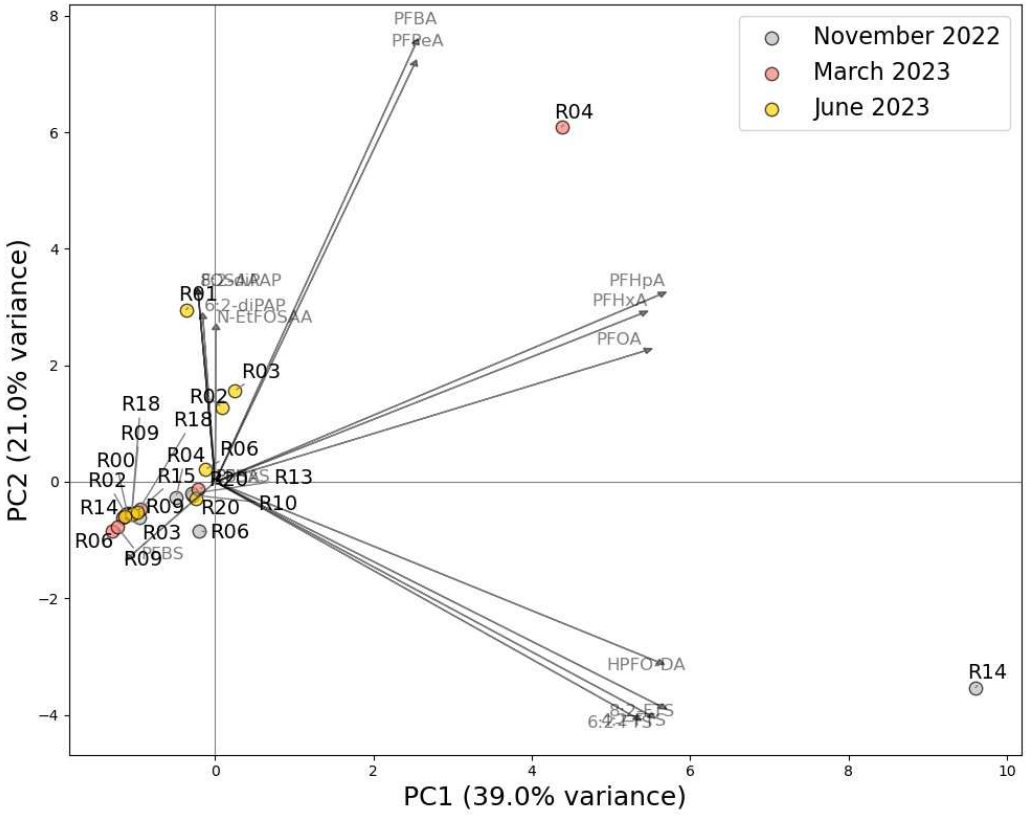

Figure 32: Principal component analysis (PCA) plot of detected PFAS in the Red River water samples.

Typically, water samples contained a wider variety of PFAS, especially short-chain compounds, while sediment tends to retain fewer PFAS, predominantly longer-chain analogues. This pattern aligns with findings by Bai et al. (2021) and Pan et al., (2014), who reported that PFAS with carbon chain lengths greater than eight (PFNA, PFDA, PFUdA, PFDoA, PFTrDA) showed higher DF in sediments compared to surface water. The contrasting PFAS profiles can be explained by the partitioning behavior, typically described using the sediment-water partitioning coefficient (K_d). Higgins et al. (2006) concluded that the carbon chain length is the primary determining factor for the sorption of PFAS into sediment, with each additional CF_2 group increasing the K_d value by a factor of 3.2 – 4.0. As a result, long-chain PFAS with lower solubility tend to sorb into sediment particles, while short-chain PFAS, characterized by low K_d values and higher mobility, preferentially remain in the aqueous phase (Okwuosa et al., 2025). This is further supported by Ahrens et al. (2009), who reported that long-chain PFAS (e.g., PFDS, and PFAS with C > 11) were exclusively found in sediments; and T. M. H. Nguyen et al. (2020) who concluded that short-chain PFAS are more frequently detected in water samples.

5.3.2. "Hidden" PFAS

TOPA was employed to investigate the contribution of oxidizable PFAS precursors in water and sediment samples from the Red River. Following oxidation, only PFAAs were detected among the 53 targeted PFAS. Following the TOPA process, there was a rise in total PFAA concentrations, partially attributed to unnoticed PFAAs in previous targeted analysis.

5.3.2.1. Sediment samples

$\sum PFAS_{sediment}$ (ng F g^{-1}) before and after TOPA in sediment samples from four sampling campaigns are shown in the *Figure 33.* Contrary to the target approach where targeted PFAS levels were mostly below the LOD, the TOPA approach revealed the additional PFAS generated from oxidizable precursors in the sediment samples. $\sum PFAS_{sediment}$ using TOPA approach ranged from n.d. – 12.0 ng g^{-1} dw in November 2022 samples (*Annex 43*), n.d. – 9.5 ng g^{-1} dw in March 2023 (*Annex 44*), 0.17 – 15 ng g^{-1} dw in June 2023 (*Annex 45*), and n.d. - 6.7 ng g^{-1} dw in September 2023 (*Annex 46*). The formation of PFAAs was evident in all sediment samples flowing oxidation. Newly formed PFCAs (C_5 – C_{10}) were consistently across all four sampling campaigns, while longer-chain PFCAs (C_{11} – C_{13}) appeared only in November 2022 samples. In addition, PFPrA (DF = 45.5%) and PFBA (DF = 18.2%) were detected in samples from March and June, respectively. PFSAs were also generated during oxidation, with PFOS detected in March and June 2023 samples and long-chain PFSAs (C_9 – C_{12}) observed in November samples 2022. The discussion on the potential precursors forming these long-chain PFAAs is presented later on.

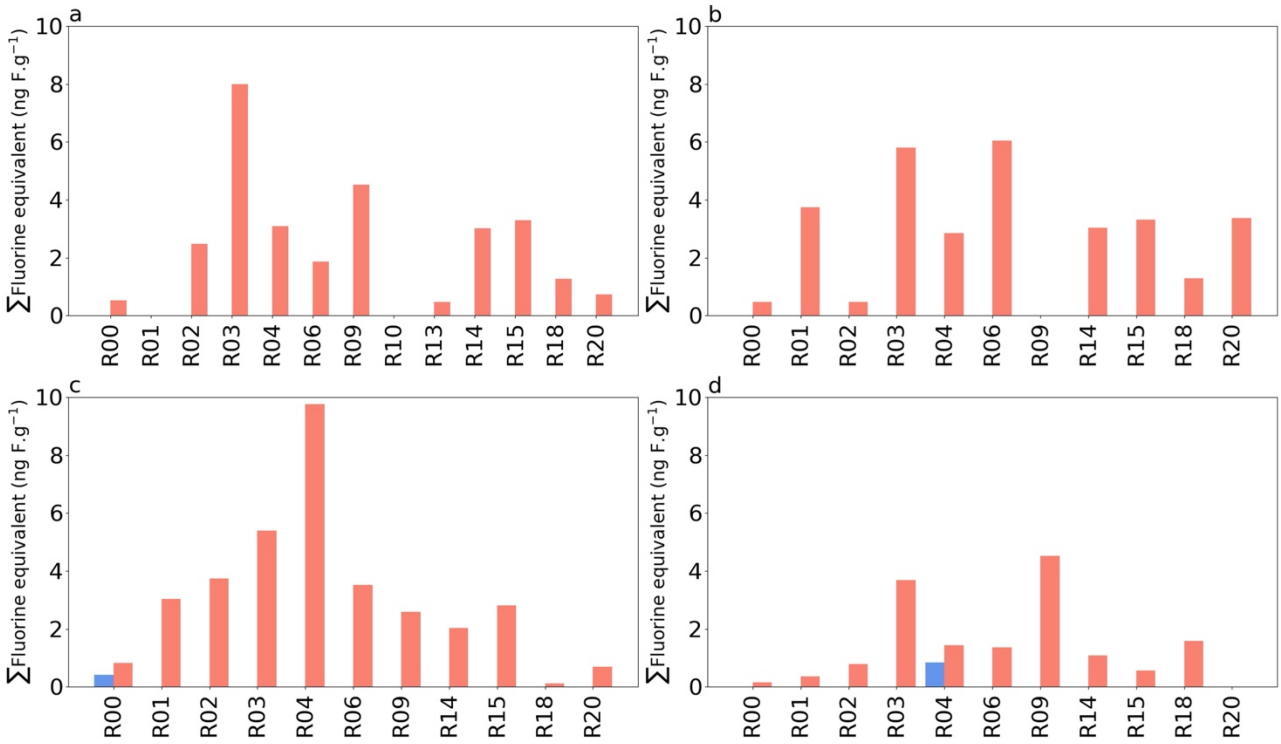

Figure 33: The comparison of F_{eq} before and after TOPA in sediment samples from: a) November 2022, b) March 2023, c) June 2023, d) September 2023

5.3.2.2. Water samples

$\sum PFAS_{water}$ (ng F L^{-1}) before and after TOPA in water samples from the three sampling campaigns are shown in the *Figure 34* and the values (in both ng L^{-1} and ng F L^{-1}) are reported in *Annex 47*, *Annex 48*, and *Annex 49*. After oxidation the $\sum PFAS_{water}$ ranged from 17.1 – 350 ng L^{-1} for November 2022 samples, 1.7 – 13.2 ng L^{-1} for March samples, and 1.6– 18.5 ng L^{-1} for June samples. PFBA, PFPeA, PFHxA, PFHpA, and PFOA are the five compounds that showed increased concentrations after the oxidation process in at least one sample across the three sampling campaigns. In November 2022, 67% of the samples exhibited higher concentrations of PFHpA and PFOA, with the target/TOPA ratio ranging from 10 to 30 and from 1.1 to 2.8, respectively. In March 2023, PFHpA was the only PFAS that increased in concentration in sample R18 (target/TOPA ratio = 2). Shorter-chain PFAS, i.e., PFBA and PFPeA, displayed increased concentrations in June samples, with changes ranging from 0.40 – 2.71 ng L^{-1} and ratios between 1.35 – 3.08.

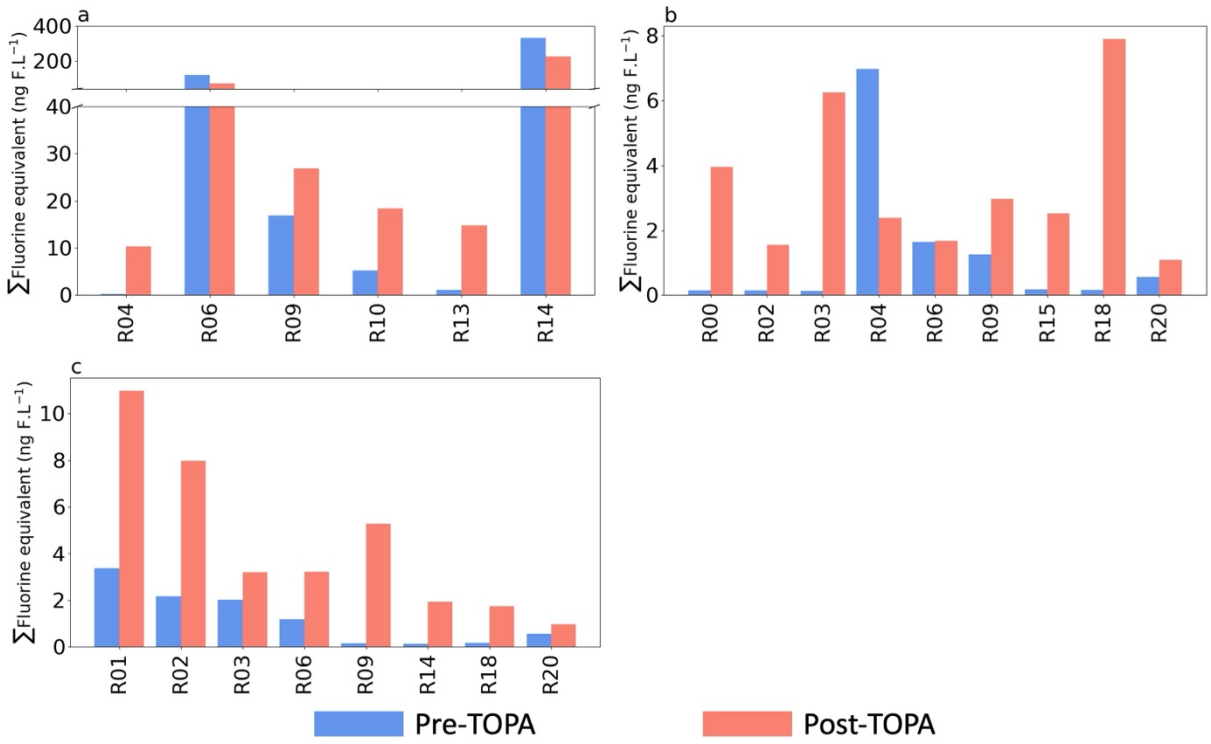

Figure 34: The comparison of F_{eq} before and after TOPA in water samples from: a) November 2022, b) March 2023, and c) June 2023 (the samples from each campaigns are not identical).

In addition to these concentration increases, several PFAAs were exclusively detected after oxidation. PFPrA was the most prevalent, occurring in over 88% of TOPA samples (June 2023). Long-chain PFAAs such as PFDA and PFUdA appeared in more than 50% of November 2022 samples and 12.5% of June 2023 samples. The TOPA samples also revealed the presence of Perfluoroalkyl sulfonic acids (PFSAs) alongside Perfluoroalkyl carboxylic acids (PFCAs). For March 2023 samples, PFBS was found in over 44% of water samples after oxidation while PFHxS, PFOS, and PFUnS were also detected but at lower DF.

5.3.2.3. PFAS burden

The concentration of individual PFAS detected in water and sediment samples before and after oxidation were converted to F_{eq} using the equation in section 2.5. The results are summarized and presented in *Table* 14 (detailed data from *Annex 43* to *Annex 49*)

Table 14: The ∑PFAS as F_{eq} in water (ng F L^{-1}) and sediment (ng F g^{-1} dw) before and after TOPA.

	November 2022		March 2023		June 2023		September 2023
	Water (n=6)	Sediment (n=13)	Water (n=9)	Sediment (n=11)	Water (n=8)	Sediment (n=11)	Sediment (n=11)
Target	0.29 – 329.90	n.d.	0.13 – 6.97	n.d.	0.14 – 3.37	n.d. – 0.41	n.d. – 0.85
TOPA	10.37 - 225.60	n.d. – 8.0	1.08 - 7.90	0 - 6.05	0.97 - 10.97	0.12 - 9.76	n.d. - 4.52
Target/TOPA (%)	60 - 3636	-	34 - 5085	-	159 - 3327	n.d. - 200	0 - 169

Even though the measured F_{eq} varied across matrices, sampling sites and campaigns, a consistent increase was generally observed after the oxidation process, indicating the presence of oxidizable PFAS precursors. In water samples, F_{eq} from target approach ranged from 0.29 to 330 ng F L^{-1}, while TOPA values were 10.37 to more than 225 ng F L^{-1}. For most samples, TOPA F_{eq} represented 150 to more than 3000% of the Feq from target approach. Three water samples (R06 and R14 in November, and R04 in March) shows the target/TOPA ratio below 100%, where target F_{eq} exceeded the values after oxidation. Among the remaining water samples, the low ratios were recorded, often below 10%, for samples in March and June 2023. On the other hand, TOPA F_{eq} in sediment samples was entirely associated with the presence of newly form PFAAs, indicating the majority of oxidizable precursors was not captured by the target approach.

The TOPA results provide clear evidence of the transformation of PFAS precursors into terminal PFAAs, highlighting the presence and the significance of both targeted and unknown oxidizable precursors in environmental samples. The fact that only PFAAs were detected in TOPA samples suggested that all targeted precursors (e.g., FTS) were effectively broken down during the oxidation process. This phenomenon was also reported in previous studies (Göckener, Lange, et al., 2022; Lange et al., 2024).

In general, the detection of C_n-PFAA after oxidation indicates the presence of corresponding C_{n+1}-precursors. Well-studied precursors (Cioni et al., 2022; Houtz & Sedlak, 2012) were identified in the Red River samples. Particularly, there was a dominance of 6:2-FTS in certain samples. This fluorotelomer, a precursor containing six fully fluorinated carbons, can be transformed into PFHxA during the oxidation process

(Antell et al., 2025). This pathway could be the explanation for the exceptional increase in PFHxA observed in water sample R14 (November 2022), where the concentration of 6:2-FTS measured using the target approach exceeded 557 ng L^{-1}, resulting in an approximate 136-fold increase. Similarly, the formation of PFHxA in water sample R06 (November 2022) further supports the contribution of 6:2-FTS and potentially other C6-precursors to the PFHxA pool in TOPA approach. Moreover, PFAS precursors could be oxidized to form PFAAs with shorter carbon chains, as previously reported by Houtz et al. (2012) and Lange et al. (2024). Houtz et al. (2012) suggested that the increase in PFHpA might arise from the oxidation of fluorotelomer-based precursors with 8 fluorinated carbons or higher such as 8:2-diPAP, 8:2-FTS, or other PFAS with 7 carbons. Therefore, the formation of PFPrA and PFBA in TOPA samples could be attributed to precursors containing 3, 4 or higher number of fluorinate carbons. This is in agreement with Martin et al. (2019) who reported that PFPrA could account for a substantial proportion of oxidation yield, up to 35%, of certain precursors, including 4:2-FTS, 6:2-FTS, and 7:3-FTCA.

In sediment samples, the detection of long-chain PFAAs after oxidation, extending up to 14 carbons, further informs the presence of long-chain precursors, with 14 or more fluorinated carbons, that were not part of the targeted compound. This finding emphasizes the limitation of the targeted analysis, as significant precursors could remain undetected, potentially leading to the underestimation of the total PFAS burden in environmental samples.

The presence of untargeted precursors was further confirmed by the calculated data on F_{eq}. The total F_{eq} of the detected PFAS in the target approach in both water and sediment samples accounted for only a small fraction of total F_{eq} after oxidation. This once again highlights that unidentified PFAS precursors might have been present in the samples and oxidized to form PFAAs.

An anomaly was observed in 3 water samples where the F_{eq} measured by TOPA exceeded the values from targeted approach, contrary to the general trend. At R04 (March 2023), the ratio target/TOPA was 34%, while the values of R06 and R14 in November were 60% and 68%, respectively. This unusual pattern can be explained by the formation of trifluoroacetic acid (TFA) during oxidation. TFA is an ultrashort chain PFAS that is rarely included in conventional target lists (Ateia et al., 2023; Janda, Odler, et al., 2019) and thus may remain undetected in routine analysis. Additionally, TFA has been proven to be highly associated with the oxidation of PFAS precursors, particularly the

fluorotelomer sulfonates (Janda et al., 2019b). Its presence in TOPA samples aligns with the carbon chain reduction mechanism mentioned earlier in the literature (Dixit et al., 2024; Göckener et al., 2022b; Müller et al., 2023), in which longer-chain PFAS precursors can be oxidized into very short-chain products. In addition to the formation after TOPA, TFA has been recently reported in environmental matrices at significant concentrations. TFA was the major PFAS in surface water from several streams in Sweden, with levels ranging from 30 to 820 ng L^{-1} (Björnsdotter et al., 2022). This ultrashort-chain PFAS was the predominant compound in groundwater, drinking water and surface water in Germany (Janda et al., 2019a) and in source water in the Philippines and Thailand (Guardian et al., 2020). Its concentration in water could reach up to several micrograms per liter, which is about 10 to 100 times higher than the other PFAS (Peter et al., 2024). Therefore, the exclusion of TFA from the target list results in notable underestimation of PFAS burden in the environmental samples.

5.4. Conclusion

This study provides the baseline data for the PFAS contamination of the downstream section of the Red River by the comprehensive assessment of PFAS by target and TOPA approaches in sediments and waters. Concentrations ranged from n.d. to 1.5 ng g^{-1} dw in sediments and from 0.2 to 572 ng L^{-1} in water samples. The detection of various PFAS compounds, including both regulated PFAAs and emerging alternatives, could reflect changing manufacturing practices and regulatory shifts. More importantly, the occurrence of these PFAS alternatives (e.g., 6:2-FTS and 6:2-diPAP) in environmental samples suggests not only their growing use but also their subsequent release into aquatic ecosystems. The environmental presence of emerging substances reflects the broader global trend of "regrettable substitution" where the phase-out of well-studied hazardous chemicals results in the adoption of structurally similar compounds that may still pose environmental and health risks but have not yet been fully evaluated or regulated.

The application of the TOPA approach provided critical insights into the hidden contribution of oxidizable PFAS precursors, revealing both increases in PFAA concentrations and the presence of new PFAAs that were not detected using the target approach. The magnitude of PFHxA formation in certain samples emphasizes the significant impact that specific precursors, especially 6:2 FTS, have on the overall PFAS profile in the water system. This highlights the importance of considering precursor

contributions when evaluating environmental PFAS contamination and potential exposure risks. TOPA enables the identification of PFAS precursors, including polyfluorinated compounds and emerging group of fluorine polymers. Finally, the consistent gap of F_{eq} between the target and TOPA approaches underscores the limitations of current target-based analysis and emphasizes the need to adapt new analysis method for the detection of TFA and to integrate non-target analysis. Such an approach would allow a more complete assessment of PFAS in environmental samples, in order to identify the untargeted compounds and minimize the underestimation of PFAS burden in the environment.

6. Conclusions and perspectives

Conclusion

The present study has answered the first specific objective and the 3 sub-objectives mentioned in the section 1.5.

Advancing the Understanding of Organic Contamination in the Red River: A large scale sampling, a screening for ubiquitous, historical, and emerging contaminants under a tripartite collaboration.

This doctoral research stands out for its original approach: it simultaneously investigates five groups of legacy and emerging organic contaminants in Vietnam's Red River, an important hydro-system within the Southeast Asian region, which is still under documented, in comparison to European countries or China. It investigates PAHs, PCBs, PBDEs, CPs, and PFAS, therefore, offering an overview of both ubiquitous historical, regulated and emerging contaminants that mostly meet the criteria of persistence, toxicity, bioaccumulation, and capability for long-range transport. PCBs, PBDEs, CPs, and PFAS hence stand out in the context of the Stockholm Convention, as they are not listed individually but rather as broad families of contaminants encompassing up to thousands of compounds. Addressing them together, along with PAHs, allowed the present work to narrow the gap between established regulatory frameworks and ongoing challenges posed by complex contaminant groups whose environmental occurrence and risks remain poorly described in Vietnam.

The scope of this work is further reinforced by the size and design of the sampling strategy. A large sample set was collected across four campaigns within one-year period, covering both dry and rainy seasons to capture hydrological variability. Moreover, the study area was extending along a 200 km transect of the Red River, covering a large deltaic zone that combines increasing urban, industrial, and agricultural influences. The large study area provides a comprehensive picture of the contamination across multiple

environmental settings, from upstream river sections to downstream zones connected to the marine environment.

Conducting this study across three laboratories, in Vietnam and France, presented methodological and logistical challenges at every stage, from sampling organization and fieldwork under changing meteorological conditions to the transport and storage of samples. The analysis of multiple groups of contaminants, each requiring specific extraction and detection methods, was based on the skills and hosting of three laboratories. This doctoral research has resulted in four research papers (one published, one accepted and, and two in preparation), with the key findings summarized and potential avenues for future research and development in the following paragraphs.

Learning from three decades of PAHs research in Vietnam and POPs studies in Southeast Asia

Research over the past three decades has demonstrated that PAHs have been widely distributed in sediments across Vietnam. Elevated concentrations have been consistently reported in urban and industrial waterways, particularly in Hanoi and Ho Chi Minh City, the two largest metropolitan areas of the country, whereas lower levels are observed in rural and coastal zones. In the majority of the studies, medium-to-high molecular weight PAHs are dominant, indicating the influence of pyrogenic sources and their persistence in sediments. While sediment quality guidelines have been well established, comprehensive monitoring programs and stricter management measures are still lacking.

PCBs, PBDEs, CPs, and PFAS are four groups of industrial chemicals of the Stockholm Convention which are not listed as individual compounds but as families of contaminants containing numerous molecules. The review of these four POP families in Southeast Asia's aquatic environments reveals a significant and enduring challenges in their environmental management. Despite the efforts to achieve the goals set by Stockholm Convention, the concentration of these POPs in the region's water resources have not shown a clear declining trend for the last 30 years. This persistence can be attributed to historical contamination and long-range transport in the case of PCBs, as well as the ongoing release of PBDEs, CPs, and PFAS from existing sources. The presence of POPs in urban waterways, marine environments, in aquatic organisms, and even tap and drinking water highlights the potential for human exposure through consumption of contaminated water and food.

The scientific literature for Southeast Asia, although growing, remains characterized by significant geographical and compound-specific knowledge gaps. For instance, a complete absence of data on CPs was reported in aquatic environments of all 11 countries, probably due to the specificity of the analysis method. Moreover, no scientific studies on the four chemical groups were found in three countries (Brunei, Myanmar, and Timor-Leste). This is undoubtedly due to the complexity of the socio-economic situations in each of these countries.

In conclusion, while Southeast Asian countries have demonstrated increasing attention to environmental quality, a strengthened and more consistent monitoring approach of POPs is critically required. Future research should focus on filling existing data gaps, particularly for CPs and monitoring efforts in three absent countries, in order to improve the understanding of the regional contamination picture. Furthermore, given the alarming factors of rapid urbanization and climate change-driven phenomenon such as intensive rainfall and flooding, further research is essential to inform policy decisions and management strategies that will ensure the long-term sustainability and the well-being of the region's water resources. In this regard, Vietnam, particularly the Red River basin, offers the representative example to explore those challenges in greater detail. The following section presents the main conclusion

PAHs in the Red River sediment (Vietnam) have highest concentration in urban zones and are mainly of pyrogenic sources, increasing during rainy season, but exhibiting low ecological risks

The total sediment concentrations of 16 priority PAHs ranged from 16.2 to 57.9 ng g^{-1} dw in Red River sediment samples collected during the dry season (March 2023). PAHs were found to be higher in samples from the rainy season from 24.5 (R12) to 94.7 (R04) ng g^{-1} dw in June and from 17.5 (R13) to 108.0 (R08) ng g^{-1} dw in September 2023. PAH concentrations were otherwise highest in urban and residential areas. For all the three periods, sediment samples displayed PAH concentrations well below Vietnam's national standards for freshwater sediments and were considered to have low to moderate pollution levels. The distribution of PAH congeners was consistent throughout the studied periods and followed the order: 4-ring > 5-ring ~ 3-ring > 6-ring > 2-ring. While Flt, Pyr, B(a)A and Chr were the most predominant PAHs, the LMW PAHs (Nap and Ace) had the lowest detection frequency in all samples. According to several diagnostic ratios, the

potential sources of PAHs the Red River sediments were mostly pyrogenic, originating from the combustion of both biomass and fossil fuels including coals, oil and petroleum. This emphasized that anthropogenic activities such as traffic and transportation, agricultural (e.g., burning crop residues) and industrial practices were significant sources of PAHs in the river. Risk assessments based on ERL/ERM, ISQG/PEL, and TEQ/MEQ approaches indicated that toxic effects of PAHs on living organisms and humans are not likely to take place.

When compared to other regions, PAH levels in the Red River are substantially lower than those reported in heavily industrialized river system of Yellow River in China (n = 61, ∑16PAHs = 403 – 16784 ng g^{-1} dw, (Z. Wang et al., 2024), Yamuna River in India (n = 5, ∑16PAHs = 4500 – 23520 ng g^{-1} dw, (Agarwal et al., 2006) . Sele River in Italy (n = 10, ∑16PAHs = 332 – 872 ng g^{-1} dw, Montuori et al., 2022). In this anthropization scale, the Red River this appears toward the lower end, despite the phase of rapid urban and industrial development being intensified since the early 2000s.

Different profiles of POPs followed their inclusion in the Stockholm Convention: Declining PCB levels, low PBDE concentrations, but rising concerns for CPs and PFAS.

The Chapter 4 provides a comprehensive investigation of three groups of POPs, i.e., PCBs, PBDEs, and CPs in the Red River sediment. By assessing their occurrence, spatial-seasonal variations, and associated ecological risks, this chapter provides new insights into the contamination issues linked to hardly degradable halogenated industrial chemicals in the Red River.

PCBs, which had been imported in Vietnam in the second half of the 20th century and have been regulated internationally since the early 2000s, were found at low concentration (n.d. – 50.5 ng g^{-1} dw). The spatial-seasonal distribution of seven indicator congeners suggested higher levels during the dry season (November 2022) compared to the rainy season (June 2023). This seasonal trend together with overall low levels compared to literature might reveal an historical contamination and a gradual decline of PCB pollution in the river, the remobilization of congeners depending on the hydrological conditions, and the effectiveness of national regulatory efforts. PBDEs, which entered the market later than PCBs but were regulated internationally in the 2010s, were also

detected at low concentrations (n.d. – 7.3 ng g^{-1} dw). Only BDE 47 and 209 were consistently detected in sediment samples in November 2022.

On the contrary, SCCPs were found at considerably higher concentrations (<LOD – 1515 ng g^{-1} dw), with a clear seasonal pattern. Their levels significantly increased during the rainy season compared to the dry season, suggesting ongoing inputs to the river. This pattern also highlights their mobility under the hydrological conditions of the Red River system. The observed increase of SCCPs during the rainy season is alarming since it reflects active contamination processes (e.g., ongoing production and usage of PVC in Vietnam) rather than historical residues as for PCBs.

Ecological risk assessment employing several guidelines showed that most samples exhibited low risks posed by both PCBs and SCCPs. However, several samples in June 2023 campaign demonstrated moderate risks associated with SCCPs. The increase in potential risk throughout time raises concerns along with the upward temporal trend of SCCP levels in the sediment. Under the hypothesis of increasing emission of CPs linked to anthropogenic pressure, ecological risk may become more significant in the near future.

The chapter 5 provides a baseline assessment of PFAS contamination in the 200km-downstream transect of the Red River. The work integrated both sediment and water samples (since most PFAS have hydrophilic properties, contrary to other studied POPs) through combined target and TOPA approaches. The 53 targeted PFAS concentrations in sediment ranged from <LOD to 1.5 ng g^{-1} dw, while in water the concentrations varied from 0.2 to 572 ng L^{-1}. The detection of multiple PFAS compounds, including both regulated PFAAs and emerging alternatives such as 6:2-FTS and 6:2-diPAP, shows evolving manufacturing practices (e.g., in food-contact packaging, papers, and cosmetics) with the substitution of regulated chemicals with structurally similar compounds. Such substitution could lead to “regrettable replacement” which emphasizes the challenge of managing emerging contaminants whose persistence, bioaccumulation potential, and ecological impacts have not yet fully understood.

Application of the TOPA approach revealed the increase in levels quantified and the formation of new PFAAs that were not detected using the target approach, which emphasized the contribution of oxidizable precursors to the total PFAS burden in the environmental samples. Moreover, the difference in fluorine equivalents before and after oxidation in both water and sediment samples indicated the limitations of the conventional target-based analysis.

In summary, the results and conclusions of this doctoral research reveal an initial advanced assessment in the Red River for five families of contaminants: the persistence of well-known regulated pollutants (e.g., PAHs, PCBs, PBDEs) and the growing presence of emerging chemicals (e.g., CPs and PFAS). PCBs can be seen as a case study of regulatory success, showing how the establishment of international and national frameworks has led to an improved control of the sources. On the contrary, CPs and PFAS represent new challenges. Their detection in the Red River underlines both their extensive use and their complex environmental behavior. Yet, their sources, contamination pathways and risk are still poorly documented in Vietnam. While the Vietnamese government has established regulations for compounds such as PAHs and PCBs, environmental guidelines for PBDEs, CPs, and PFAS are still absent or remain incomplete, leaving breaches in chemical monitoring and management.

Perspectives

Several strategies could strengthen the understanding and management of contaminant dynamics in the Red River system.

Improving the field campaign design and the regional environmental surveys

There are always limitations in field campaigns, and a first step is a step forward. However future work would benefit from field campaigns and monitoring networks being optimized.

A major first step toward improving the understanding and management of chemical contamination in the Red River basin would consist of establishing a coordinated and systematic monitoring network. Such a network should include regular (e.g., seasonally) sampling campaigns not only along the main branch of the River but also major tributaries, in order to capture both spatial and temporal variations in contaminant levels. This monitoring could be supported by a shared data platform and integrated with modelling frameworks as promoted by the Intergovernmental Science-Policy Panel on Chemicals, Waste, and Pollution (ISP-CWP). This structure would ensure data comparability and facilitate regional collaboration. While occasional, campaign-based studies are useful for identifying contamination hotspots or pointing out contaminant of

concern, they often fail to capture the temporal variability characteristic of dynamic river systems such as the Red River, where hydrological conditions, land use, and human activities can change rapidly.

To obtain a more comprehensive picture of contaminant dynamics, the monitoring framework should combine the analysis of water, sediment, and biota samples. This multi-approach would allow a better understanding of contaminant's eco-dynamic, in the typical context of tropical environments.

Direct PCB inputs into the area seem possible, but it is also likely that contamination has been brought in by long-distance transport. On the other hand, understanding the dynamics between industrial innovation, market introduction, regulatory restriction and prohibition, and phase shift. This could be done by examining the evolution of concentrations over time In this frame, , the analysis of sediment cores could provide insights into the historical evolution of contamination over decades, although this technique requires to ascertain the stratification of the sedimentation and careful core calibration and dating . Another way to evaluate decadal temporal trends is to perform opportunistic retrospective analysis on archived samples (e.g., suspended particulate matter) from partner laboratories. But this necessitates that proper sampling archiving has been carried out.

In addition to monitoring on abiotic matrices, the inclusion of biological surveillance would provide critical ecological context. The use of benchmark species (e.g., mussels, shrimps, fish, or benthic invertebrates) and biofilms (specific tropical diatom species) as bioindicators can reveal signs of bioaccumulation and biological effects that chemical data on abiotic samples cannot fully describe. This biological perspective would enable the development of ecotoxicological reference frameworks specific to tropical systems in Vietnam.

Implementing novel sampling, analytical and modelling strategies

To make sense of water analysis campaigns, it is often necessary to repeat them in order to identify trends, but this increases costs and workload. Furthermore, we did not analyze hydrophobic POPs in water (PAHs, PCB, CPs, PBDEs), because concentrations are generally below the LOD. However, this data is lacking in order to better identify sources and understand the dynamics of inputs and remobilization. The application of water

passive samplers represents a promising and complementary approach, as it enables time-weighted and cost-effective monitoring of contaminants in aquatic systems. Passive samplers reduce the logistical constraints associated with frequent field campaigns and provide data that are often more representative of environmental exposure than campaign-based on spot sampling. However, the use of these tools for certain molecules considered in my work, such as PFAS in particular, requires further in-lab complex methodological developments. The data on occurrence in water provided by my research forms one of the bases for defining the list of molecules to target in order to develop such an approach.

My thesis enabled the implementation of two methods, one targeted and one more comprehensive, (TOPA) for quantifying PFAS. It highlighted a discrepancy between target and TOPA quantification highlighting a gap in the identification of environmental fluorinated compounds. But we simply could put forward hypotheses to justify these discrepancies. This issue needs to be explored further, in particular by combining it with non-targeted analysis. This could be performed for instance on samples having large difference in results from both methods and on sediment specifically as they exhibit fluorine content, but no PFAS could have been detected. Another hypothesis which must be explored is the search/screening for the fluorinated polymers, which are not currently studied in the environment, neither in molecular compounds studies nor in microplastic studies. On the other hand, the analytical approach used for CPs analysis is also a global method enabling the rapid screening of environmental samples for the presence of CPs. If comprehensive, it does not enable for instance neither to discriminate between SCCPs and MCCPs nor to obtain a fine view of the congener profiles. However, this is of interest when seeking to distinguish between sources or study the dynamics between regulation and the chemical substances market. So, the sampling area with positive detection would benefit for the implementation of method based on GC/HRMS for congeners' profile characterization.

Furthermore, the integration of hydrological and environmental models with chemical monitoring data would allow the quantification of i) contaminant fluxes from inland areas to the marine environment via riverine transport, and ii) contaminants dispersion within the river and in the adjacent coastal environment. In this context, the sediment data collected in this study, specially, concentrations of POPs across multiple campaigns, particle size distribution, and TOC content, would provide critical inputs for

parameterizing contaminant partitioning and sediment transport within such models. For instance, particle size and TOC can inform contaminant sorption dynamics, while temporal concentration data allow model validation against observed seasonal trends. However, additional data such as river discharge, hydrodynamic conditions, and upstream contaminant sources would be needed to fully implement these models. Linking the current dataset with these missing parameters would make it possible to estimate source contributions, seasonal fluxes, and transport pathways, providing a concrete basis for targeted mitigation and policy decisions.

From developing countries to sustainable developing countries

On a broader global perspective, the findings of this thesis highlight not only local environmental challenges but also issues at the intersection of economic development, social equity, climate change, and chemical management. The methodological approaches, combining targeted and global analyses, spatial mapping, and multi-compartment and multi-chemical monitoring, developed in this doctoral work can be transferred to other tropical river systems, notably in Southeast Asia. Applying these methods more broadly could enhance regional cooperation, improve comparability of data, and support evidence-base policy decisions aimed at mitigating the environmental and social impacts of POPs.

Another major issue concerns waste management and the circular economy. Inadequate treatment facilities, informal recycling and open burning of waste contribute the release of POPs. These operations usually occur in poorly regulated conditions, without proper safety measures. Therefore, workers, usually from lower-income or marginalized communities, and nearby residents are exposed to hazardous contaminants. This exposes a pattern of environmental inequality, in which socially and economically disadvantaged people bear a disproportionate share of pollution burdens. Protecting these vulnerable populations, especially workers, children, and rural communities, would constitute an ethical imperative and an effective environmental strategy, as improving working and living conditions could lead to wider ecosystem protection by controlling and limiting the release of hazardous waste to the environment. To that end, the development of human monitoring programs using non-invasive indicators (such as hair and urine analysis) could be particularly valuable for identifying exposure pathways and

assessing cumulative risks to human health. This assessment would strengthen the link between environmental, occupational and public health, and could lead to the improvement of labor and environmental protection policies and the implementation of "One Health" approach as proposed by WHO.

Finally, Vietnam's history underscores the importance of learning from past contamination crises. The long-lasting legacy of Agent Orange demonstrates a painful yet valuable lesson on how large-scale, poorly managed chemical use could leave behind intergenerational scars on both ecosystem and human health. In this regard, the contemporary challenge of managing organic contaminants and emerging POPs must be seen as a part of broader effort to avoid repeating past mistakes while ensuring that economic growth and development do not come at the expense of environmental and social sustainability.

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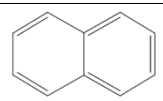
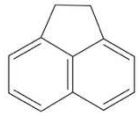
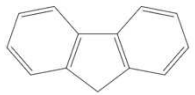
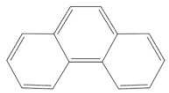
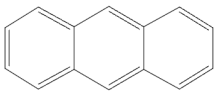
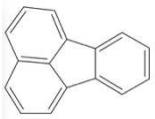
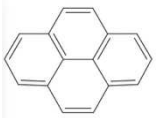
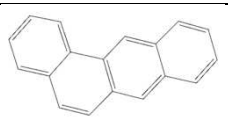
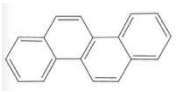
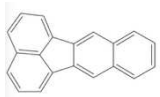
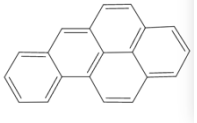
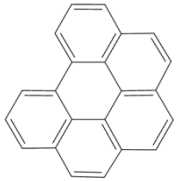
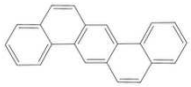
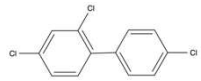
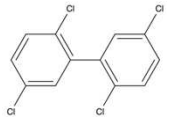
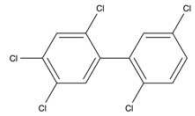
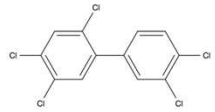
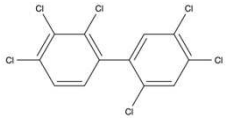
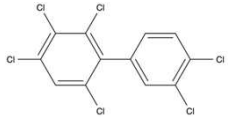
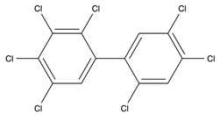
Annex

Annex 1: Sixteen priority PAHs

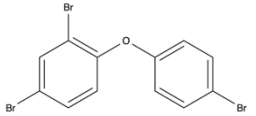
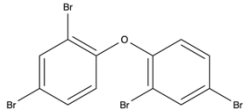
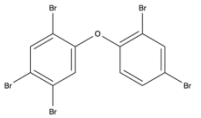
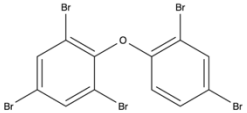
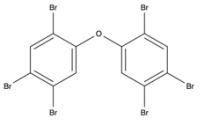
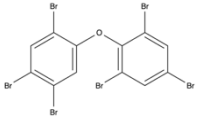
Name	Acronym	Cas number	Formula	Molar Mass (g mol^{-1})	logK_{ow}	Chemical structure
Naphthalene	Nap	91-20-3	$C_{10}H_8$	128.17	3.3	
Acenaphthylene	Acy	208-96-8	$C_{12}H_8$	152.19	3.7	
Acenaphthene	Ace	83-32-9	$C_{12}H_{10}$	154.21	3.9	
Fluorene	Flu	86-73-7	$C_{13}H_{10}$	166.22	4.2	
Phenanthrene	Phe	85-01-8	$C_{14}H_{10}$	178.23	4.5	
Anthracene	Ant	120-12-7	$C_{14}H_{10}$	178.23	4.4	
Fluoranthene	Flt	206-44-0	$C_{16}H_{10}$	202.25	5.2	
Pyrene	Pyr	129-00-0	$C_{16}H_{10}$	202.25	4.9	

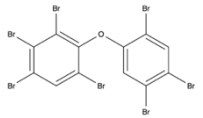
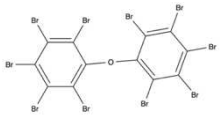
Benzo(a)Anthracene	BaA	56-55-3	$C_{18}H_{12}$	228.3	5.8	
Chrysene	Chr	218-01-9	$C_{18}H_{12}$	228.3	5.7	
Benzo(b)Fluoranthene	BbF	205-99-2	$C_{20}H_{12}$	252.3	6.4	
Benzo(k)Fluoranthene	BkF	207-08-9	$C_{20}H_{12}$	252.3	6.4	
Benzo(a)Pyrene	BaP	50-32-8	$C_{20}H_{12}$	252.3	6.0	
Indeno(1,2,3-c,d)Pyrene	Ind	193-39-5	$C_{22}H_{12}$	276.3	7.0	
Benzo(g,h,i)Perylene	BghiP	191-24-2	$C_{22}H_{12}$	276.6	6.6	
diBenzo(a,h)Anthracene	DBahA	53-70-3	$C_{22}H_{14}$	278.3	6.5	

Annex 2: Seven indicator PCBs

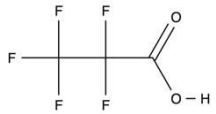
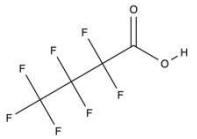
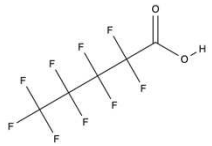
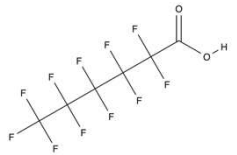
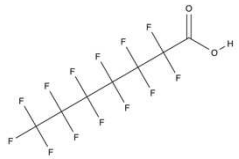
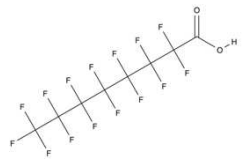
IUPAC name	Congener Number	Cas number	Formula	Molar Mass (g mol^{-1})	logK_{ow}	Chemical structure
2,4,4'-Trichlorobiphenyl	28	7012-37-5	$C_{12}H_7Cl_3$	257.5	5.6	
2,2',5,5'-Tetrachlorobiphenyl	52	35693-99-3	$C_{12}H_6Cl_4$	292.0	6.1	
2,2',4,5,5'-Pentachlorobiphenyl	101	37680-73-2	$C_{12}H_5Cl_5$	326.4	6.5	
2,3',4,4',5-Pentachlorobiphenyl	118	31508-00-6	$C_{12}H_5Cl_5$	326.4	7.1	
2,2',3,4,4',5'-Hexachlorobiphenyl	138	35065-28-2	$C_{12}H_4Cl_6$	360.9	7.2	
2,3,3',4,4',6-Hexachlorobiphenyl	158	74472-42-7	C12H4Cl6	360.9	7.2	
2,2',3,4,4',5,5'-Heptachlorobiphenyl	180	35065-29-3	C12H3Cl7	395.3	7.9	

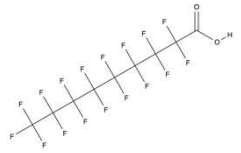
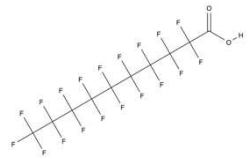
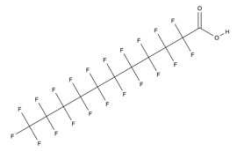
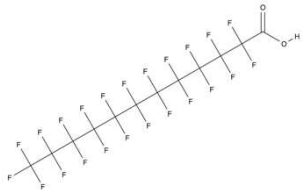
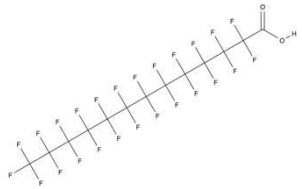
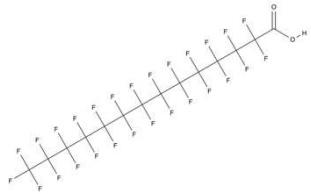
Annex 3: Eight targeted PBDEs

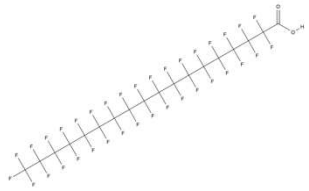
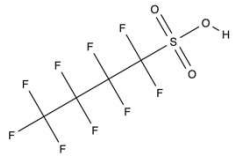
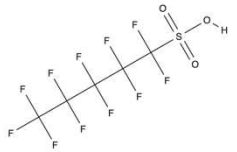
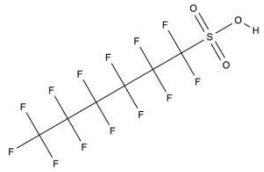
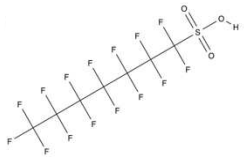
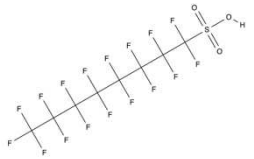
IUPAC name	Congener Number	Cas number	Formula	Molar Mass (g mol^{-1})	logK_{ow}	Chemical structure
2,4,4'-tribromodiphenyl ether	28	41318-75-6	$C_{12}H_7Br_3O$	406.89	5.5	
2,2',4,4'-tetrabromodiphenyl ether	47	5436-43-1	$C_{12}H_6Br_4O$	485.79	6.2	
2,2',4,4',5-pentabromodiphenyl ether	99	60348-60-9	$C_{12}H_5Br_5O$	564.7	6.9	
2,2',4,4',6-pentabromodiphenyl ether	100	189084-64-8	$C_{12}H_5Br_5O$	564.7	6.9	
2,2',4,4',5,5'-hexabromodiphenyl ether	153	68631-49-2	$C_{12}H_4Br_6O$	643.6	7.6	
2,2',4,4',5,6'-hexabromodiphenyl ether	154	207122-15-4	$C_{12}H_4Br_6O$	643.6	7.6	

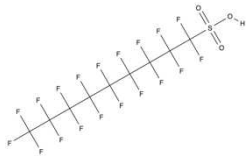
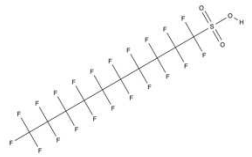
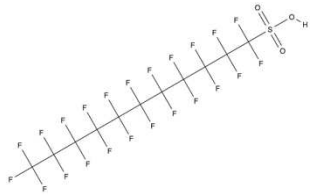
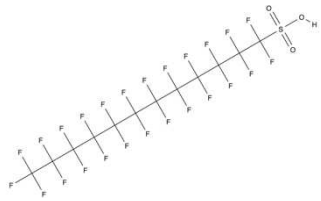
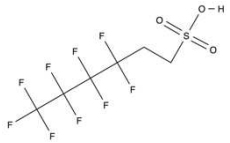
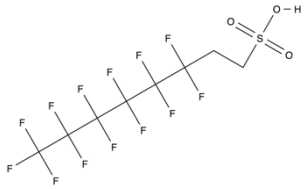
2,2',3,4,4',5',6-heptabromodiphenyl ether	183	207122-16-5	$C_{12}H_3Br_7O$	722.5	8.3	
Decabromodiphenyl ether	209	1163-19-5	$C_{12}Br_{10}O$	959.2	10.4	

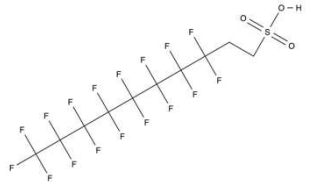
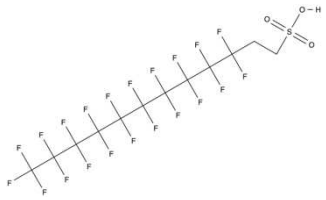
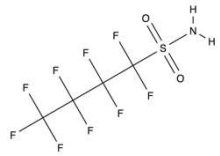
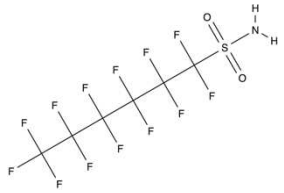
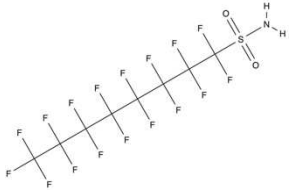
Annex 4: Fifty-three targeted PFAS

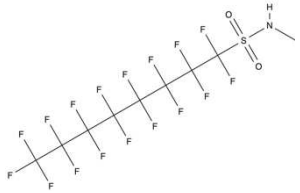
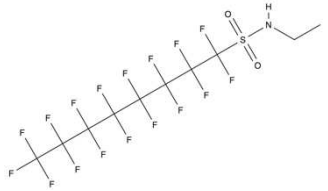
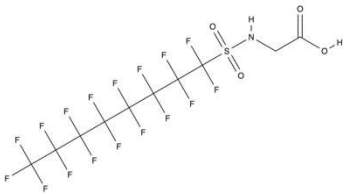
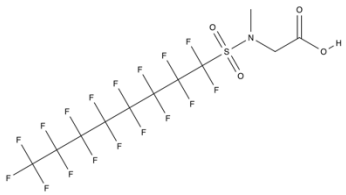
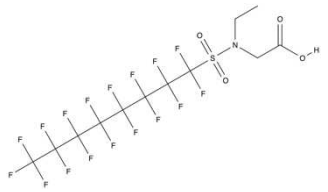
Name	Acronym	Cas Number	Formula	Molar mass (g mol^{-1})	logK_{ow}	Chemical structure
Perfluoropropionic acid	PFPrA	422-64-0	$C_3HF_5O_2$	164.03	1.6	
Perfluorobutanoic acid	PFBA	375-22-4	$C_4HF_7O_2$	214.04	2.2	
Perfluoropentanoic acid	PFPeA	2706-90-3	$C_5HF_9O_2$	264.05	2.9	
Perfluorohexanoic acid	PFHxA	307-24-4	$C_6HF_{11}O_2$	314.05	3.6	
Perfluoroheptanoic acid	PFHpA	375-85-9	$C_7HF_{13}O_2$	364.06	4.3	
Perfluorooctanoic acid	PFOA	335-67-1	$C_8HF_{15}O_2$	414.07	4.9	

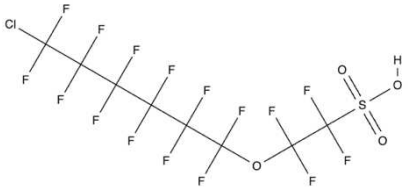
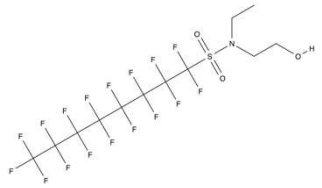
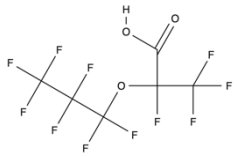
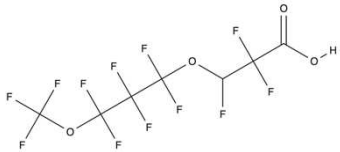
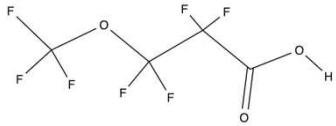
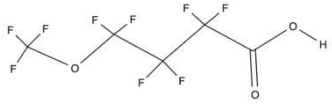
Perfluorononanoic acid	PFNA	375-95-1	$C_9HF_{17}O_2$	464.08	5.6	
Perfluorodecanoic acid	PFDA	335-76-2	$C_{10}HF_{19}O_2$	514.08	6.3	
Perfluoroundecanoic acid	PFUdA	2058-94-8	$C_{11}HF_{21}O_2$	564.09	6.9	
Perfluorododecanoic acid	PFDoA	307-55-1	$C_{12}HF_{23}O_2$	614.1	7.6	
Perfluorotridecanoic acid	PFTrDA	72629-94-8	$C_{13}HF_{25}O_2$	664.10	8.3	
Perfluorotetradecanoic acid	PFTeDA	376-06-7	$C_{14}HF_{27}O_2$	714.11	9	

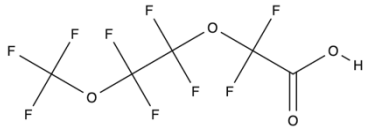
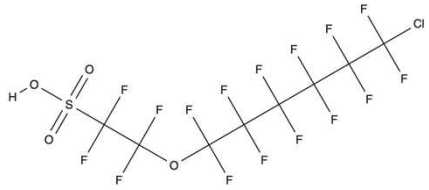
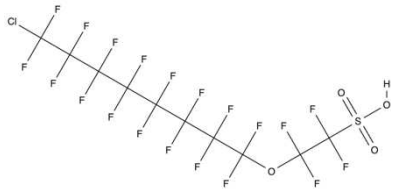
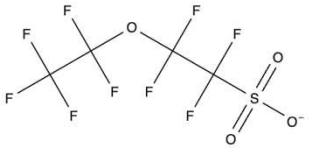
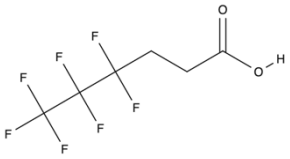
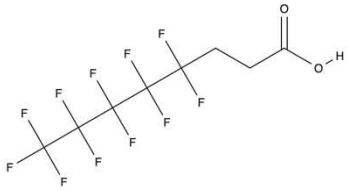
Perfluorooctadecanoic acid	PFODA	16517-11-6	$C_{18}HF_{35}O_2$	914.1	11.6	
Perfluorobutanesulfonic acid	PFBS	375-73-5	$C_4HF_9O_3S$	300.10	2.3	
Perfluoropentanesulfonic acid	PFPeS	2706-91-4	$C_5HF_{11}O_3S$	350.11	3	
Perfluorohexanesulfonic acid	PFHxS	355-46-4	$C_6HF_{13}O_3S$	400.12	3.7	
Perfluoroheptanesulfonic acid	PFHpS	375-92-8	$C_7HF_{15}O_3S$	450.12	4.3	
Perfluorooctanesulfonic acid	PFOS	1763-23-1	$C_8HF_{17}O_3S$	500.13	5.0	

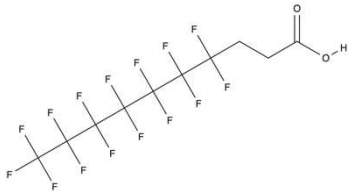
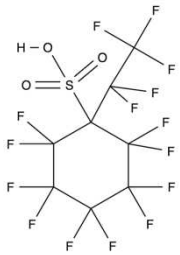
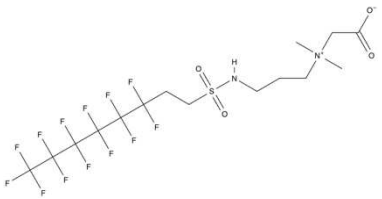
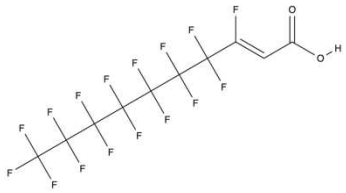
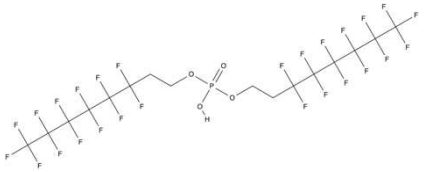
Perfluorononanesulfonic acid	PFNS	68259-12-1	$C_9HF_{19}O_3S$	550.14	5.7	
Perfluorodecanesulfonic acid	PFDS	335-77-3	$C_{10}HF_{21}O_3S$	600.15	6.4	
Perfluoroundecanesulfonic acid	PFUnS	749786-16-1	$C_{11}HF_{23}O_3S$	650.15	7	
Perfluorododecanesulfonic acid	PFDoS	79780-39-5	$C_{12}HF_{25}O_3S$	700.16	7.7	
4:2 fluorotelomer sulfonic acid (1H,1H,2H,2H-Perfluorohexanesulfonic acid)	4:2-FTS	757124-72-4	$C_6H_5F_9O_3S$	328.15	2.6	
6:2 fluorotelomer sulfonic acid (1H,1H,2H,2H-perfluorooctanesulfonic acid)	6:2-FTS	27619-97-2	$C_8H_5F_{13}O_3S$	428.17	3.9	

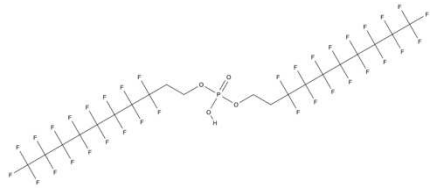
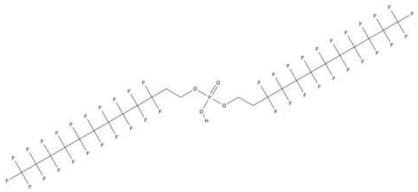
8:2 fluorotelomer sulfonic acid (1H,1H,2H,2H-perfluorodecanesulfonic acid)	8:2-FTS	39108-34-4	$C_{10}H_5F_{17}O_3S$	528.18	5.3	
10:2 fluorotelomer sulfonic acid	10:2-FTS	120226-60-0	$C_{12}H_5F_{21}O_3S$	628.20	6.6	
Perfluorobutanesulfonamide	FBSA	30334-69-1	$C_4H_2F_9NO_2S$	299.12	2.2	
Perfluorohexanesulfonamide	FHxSA	41997-13-1	$C_6H_2F_{13}NO_2S$	399.13	3.5	
Perfluorooctanesulfonamide	FOSA	754-91-6	$C_8H_2F_{17}NO_2S$	499.15	4.8	

N-Methylperfluoro-1-octanesulfonamide	N-MeFOSA	31506-32-8	$C_9H_4F_{17}NO_2S$	513.17	5.2	
N-Ethylperfluoro-1-octanesulfonamide	N-EtFOSA	4151-50-2	$C_{10}H_6F_{17}NO_2S$	527.2	5.6	
Perfluorooctane sulfonamidoacetic acid	FOSAA	2806-24-8	$C_{10}H_4F_{17}NO_4S$	557.18	4.8	
2-(N-Methylperfluorooctane sulfonamido)acetic acid	N-MeFOSAA	2355-31-9	$C_{11}H_6F_{17}NO_4S$	571.21	5	
N-Ethylperfluorooctane sulfonamidoacetic acid	N-EtFOSAA	2991-50-6	$C_{12}H_8F_{17}NO_4S$	585.24	5.4	

N-Methylperfluorooctane sulfonamidoethanol	N-MeFOSE	24448-09-7	$C_{11}H_8F_{17}NO_3S$	557.23	4.7	
N-Ethylperfluorooctane sulfonamidoethanol	N-EtFOSE	1691-99-2	$C_{12}H_{10}F_{17}NO_3S$	571.25	5.1	
2,3,3,3-tetrafluoro-2-(heptafluoropropoxy)propionic acid GenX	HPFO-DA	13252-13-6	$C_6HF_{11}O_3$	330.05	3.6	
Perfluoro-4,8-dioxa-3H-nonanoic acid	ADONA	919005-14-4	$C_7H_2F_{12}O_4$	378.07	4.1	
Perfluoro-4-oxapentanoic acid	PF4OPeA	377-73-1	$C_4HF_7O_3$	230.04	2.3	
Perfluoro-5-oxahexanoic acid	PF5OHxA	863090-89-5	$C_5HF_9O_3$	280.04	3	

Perfluoro-3,6-dioxaheptanoic acid	3,6-OPFHpA	151772-58-6	$C_5HF_9O_4$	296.04	3	
9-chlorohexadecafluoro-3-oxanone-1-sulfonic acid	9Cl-PF3ONS	756426-58-1	$C_8HClF_{16}O_4S$	532.58	5.3	
11-Chloroperfluoro-3-oxoundecane sulfonic acid	11Cl-PF3OUdS	763051-92-9	$C_{10}HClF_{20}O_4S$	632.6	6.7	
Perfluoro(2-ethoxyethane)sulfonate	PFEESA	113507-82-7	$C_4HF_9O_4S$	316.10	2.4	
3-Perfluoropropyl propanoic acid	3:3-FTCA	356-02-5	$C_6H_5F_7O_2$	242.09	2.5	
3-Perfluoropentyl propanoic acid	5:3-FTCA	811-572-3	$C_8H_5F_{11}O_2$	342.11	3.9	

3-Perfluoroheptyl propanoic acid	7:3-FTCA	812-70-4	$C_{10}H_5F_{15}O_2$	442.12	5.2	
Perfluoroethylcyclohexane sulfonic acid	PFECHS	1135682-88-0	$C_8HF_{15}O_3S$	462.13	3.9	
6:2 fluorotelomer sulfonamide betaine	6:2-FTAB	34455-29-3	$C_{15}H_{19}F_{13}N_2O_4S$	570.4	4.8	
2H-Perfluoro-2-decenoic acid	8:2-FTUCA	70887-84-2	$C_{10}H2F_{16}O_2$	458.10	5.1	
6:2 Fluorotelomer phosphate diester	6:2-diPAP	975-653-6	$C_{16}H_9F_{26}O_4P$	790.17	8.6	

8:2 Fluorotelomer phosphate diester	8:2-diPAP	678-41-1	$C_{20}H_9F_{34}O_4P$	990.2	11.3	
10:2 Fluorotelomer phosphate diester	10:2-diPAP		$C_{24}H_9F_{42}O_4P$	1190.2		

Annex 5: National regulations and status of ratification of the Stockholm Convention in some Southeast Asian Countries

Countries	National Regulations	Stockholm Convention
Brunei Darussalam	Hazardous Waste Order (2013) Environmental Protection and Management order (2016)	Signature* in 2002
Cambodia	Water pollution control (1999, amended in 2021) Drinking Water Quality Standards (2004)	Ratification in 2006
Indonesia	Government Regulation No. 74, 2001 on hazardous and toxic substances management Ministerial Regulation P.29/MENLHK/SETJEN/PLB.3/12/2020 on the Management of Hazardous waste	Ratification in 2009
Laos	Decision No. 5925/MONRE on the Management, Monitoring, Treatment, and Disposal of PCBs	Ratification in 2006
Malaysia	Environmental quality regulations (2009)	Signature** in 2002
Myanmar	Environmental Conservation Act No. 9/2012	Ratification in 2004
The Philippines	Chemical Control Orders Administrative order No.18A.1997 on Standards of Quality and Requirements for the Processing, Packaging and Labeling of Bottled Drinking Water	Ratification in 2004
Singapore	Environmental Protection and Management Act 1999 (revised in 2021) Hazardous Waste Act 1997 (amended 2003) Environmental Public Health (Water Suitable for Drinking) Regulations 2019	Ratification in 2005
Thailand	Hazardous Substances Act Ministerial Regulation Soil and Groundwater Contamination Control in Factory Area B.E. 2559 (2016)	Ratification in 2005

	Notification of the Department of Health on Critearia for Recommendation of Drinking Water Quality Surveillance B.E. 2563 (2020)	
Timor-Leste	Environmental Basic Legislation (Decree-Law No. 26/2012)	-
Vietnam	QCVN 08:2023/BTNMT: National technical regulation on surface water quality QCVN 43:2017/BTNMT: National technical regulation on sediment quality Law on Chemicals and degree 82/2022/ND-CP : Restriction in the usage of PCBs, decaBDEs, PFOA and PFHxS and short-chain CPs in indutries	Ratification 2002

*Ratification is the international act whereby State establishes on the international plane its consent to be bound by a treaty (United Nations, 1969).

**Signature is a means of authentication and expresses the willingness of the signatory state to continue the treaty-making process (United Nations, 1969).

Annex 6: Articles investigating the occurrence of PCBs, PBDEs and PFAS in 8 Southeast Asian countries

	No	Article title	Analytes	Samples	Concentration	Reference
Cambodia	1	Persistent Organochlorine Residues in Marine and Freshwater Fish in Cambodia	PCBs	48 freshwater fish	< 0.05– 1 ng g^{-1} ww	(Monirith et al., 1999)
				33 marine fish	0.07 – 1.2 ng g^{-1} ww	
	2	Organochlorine contamination in fish and mussels from Cambodia and other Asian countries	PCBs (concentrations of individually resolved peak areas relative to an equivalent mixture standard of Aroclor (1016:1242:1254:1260; 1:1:1:1)	Green mussels *(Perna viridis)*	< 0.05 – 5.1 ng g^{-1} ww	(Monirith et al., 2000)
	3	Asia-Pacific mussel watch: monitoring contamination of persistent organochlorine compounds in coastal waters of Asian countries	PCBs (concentrations of individually resolved peak areas relative to an equivalent mixture standard of Aroclor (1016:1242:1254:1260; 1:1:1:1)	Green mussels *(Perna viridis)*	< 3.8 – 220 ng g^{-1} lw	(Monirith et al., 2003)
	4	Asian Mussel Watch Program: Contamination Status of Polybrominated Diphenyl Ethers and Organochlorines in Coastal Waters of Asian Countries	62 PCBs	Mussels	200 - 300 ng g^{-1} lw	(Ramu et al., 2007)
			14 PBDEs	Mussels	2.3 – 5.3 ng g^{-1} lw	

	5	Sedimentary PBDEs in urban areas of tropical Asian countries	23 PBDEs	Sediment in urban canals, rivers and marine bay	2.13 – 652 ng g^{-1} dw	(Kwan, Takada, Mizukawa, Saha, et al., 2013)
	6	PBDEs in leachates from municipal solid waste dumping sites in tropical Asian countries: phase distribution and debromination	46 PBDEs	Leachates from solid waste dumping sites	308 – 369 ng L^{-1}	(Kwan, Takada, Mizukawa, Torii, et al., 2013)
Indonesia	1	Geographical distribution of Persistent Organochlorines in air, water and sediments from Asia and Oceania, and their implications for global redistribution from lower latitudes	PCBs	River water from urban and rural zones	0.38 – 2.1 ng L^{-1}	(Iwata et al., 1994)
				River sediment from urban zones	5.9 – 220 ng g^{-1} dw	
	2	Organochlorine contamination in fish and mussels from Cambodia and other Asian countries	PCBs (concentrations of individually resolved peak areas relative to an equivalent mixture standard of Aroclor (1016:1242:1254:1260; 1:1:1:1)	Green mussels *(Perna viridis)*	0.2 – 2.7 ng g^{-1} ww	(Monirith et al., 2000)
	3	Asia-Pacific mussel watch: monitoring contamination of persistent	PCBs (concentrations of individually resolved peak areas relative to an equivalent mixture standard of Aroclor	Green mussels *(Perna viridis)*	5.6 – 210 ng g^{-1} lw	(Monirith et al., 2003)

		organochlorine compounds in coastal waters of Asian countries	(1016:1242:1254:1260; 1:1:1:1)			
	4	Global pollution monitoring of PCBs and Organochlorine Pesticides using Skipjack Tuna as a Bioindicator	117 PCB detected	Liver of Skipjack Tuna (*Katsuwonus pelamis)*	42 – 58 ng g^{-1} lw	(Ueno et al., 2003)
	5	Global pollution monitoring of Polybrominated Diphenyl Ethers Using Skipjack Tuna as a Bioindicator	11 PBDEs	Muscle tissue (pooled samples) of Skipjack Tuna (*Katsuwonus pelamis)*	3.1 ng g^{-1} lw	(Ueno et al., 2004)
	6	Asian Mussel Watch Program: Contamination Status of Polybrominated Diphenyl Ethers and Organochlorines in Coastal Waters of Asian Countries	62 PCBs	Mussels	510 - 1400 ng g^{-1} lw	(Ramu et al., 2007)
			14 PBDEs	Mussels	12.38 – 13.68 ng g^{-1} lw	
	7	Levels and distribution of organochlorines in fish from Indonesia	62 PCBs	Pooled whole body homogenized samples of 25 species of marine and freshwater fish	9.7 – 2700 ng g^{-1} lw	(Sudaryanto et al., 2007)

	8	Characterization of polychlorinated biphenyls and brominated flame retardants in sediments from riverine and coastal waters of Surabaya, Indonesia	62 PCBs	River sediment	0.07 – 420 ng g^{-1} dw	(Ilyas et al., 2011)
				Marine sediment	< LOD – 0.75 ng g^{-1} dw	
			14 PBDEs	River sediment	0.53 – 35.5 ng g^{-1} dw	
				Marine sediment	< LOD – 3.04 ng g^{-1} dw	
	9	Perylene dominates the organic contaminant profile in the Berau delta, East Kalimantan, Indonesia	32 PCBs	River and marine sediment	< LOD	(Booij et al., 2012)
	10	Distribution, source identification, and historical trends of organic micropollutants in coastal sediment in Jakarta Bay, Indonesia	26 PCBs	Coastal sediment	2 – 9 ng g^{-1} dw	(Rinawati et al., 2012)
	11	Characterization of polychlorinated biphenyls and brominated flame retardants in sludge, sediment and fish from municipal dumpsite at Surabaya, Indonesia	62 PCBs	Sediment and sludge from municipal dumpsite	n.d. – 60 ng g^{-1} dw	(Ilyas et al., 2013)
				Pooled fish sample	30 – 55 ng g^{-1} lw	
			14 PBDEs	Sediment and sludge from municipal dumpsite	0.0075 – 45 ng g^{-1} dw	

				Pooled fish sample	6.6 – 11 ng g^{-1} lw	
	12	Sedimentary PBDEs in urban areas of tropical Asian countries	23 PBDEs	Sediment in urban canals, rivers and marine bay	11.5 - 202 ng g^{-1} dw	(Kwan, Takada, Mizukawa, Saha, et al., 2013)
	13	PBDEs in leachates from municipal solid waste dumping sites in tropical Asian countries: phase distribution and debromination	46 PBDEs	Leachates from solid waste dumping sites	499 ng L^{-1}	(Kwan, Takada, Mizukawa, Torii, et al., 2013)
	14	Water quality status of Segara Anakan Cilacap Indonesia for Biota Life	PCBs	Estuarine water	68 – 83 ng L^{-1}	(Piranti et al., 2020)
	15	Evidence of Micropollutants in Sediment and Mud Clams (*Polymesoda erosa*) from One of Mangrove Biodiversity Hotspots in Indonesia	PCB-28, -52, -101	Lagoon sediment	< LOD – 2.63 ng g^{-1} dw	(Yudhistira Azis et al., 2021)
			7 PCBs	Mud clam (*Polymesoda erosa*)	2.34 ng g^{-1} tissue weight	
	16	First insight into accumulation of characteristics and tissues distribution of PCBs, PBDEs, and other BFRs in the living Indonesian coelacanth (*Latimeria menadoensis*)	209 PCBs	Indonesian coelacanth (*Latimeria menadoensis*)	Liver: 120 ng g^{-1} lw Muscle: 86 ng g^{-1} lw Ovary: 15 ng g^{-1} lw	(Falahudin et al., 2022)
			PBDEs		Liver: 3.9 ng g^{-1} lw	

			(9 detected in liver, 3 in muscle and only BDE-47 in ovary)		Muscle: 6.1 ng g^{-1} lw Ovary: 4.5 ng g^{-1} lw	
	17	The first full-congener analysis of 209 polychlorinated biphenyls (PCBs) in the blubber of short-finned pilot whales (*Globicephala macrorhynchus)* stranded along the coast of Savu Island, Indonesia	209 PCBs	Short-finned pilot whales (*Globicephala macrorhynchus)*	47 – 480 ng g^{-1} lw	(Falahudin et al., 2023)
Laos	1	Assessment of persistent organic pollutants in sediments from Lower Mekong River Basin	62 PCBs	Mekong River sediment	0.21 – 7.8 ng g^{-1} dw	(Sudaryanto et al., 2011)
				Sediment from canals in Vientiane	11 -300 ng g^{-1} dw	
	2	Sedimentary PBDEs in urban areas of tropical Asian countries	23 PBDEs	Sediment in urban canals, rivers and marine bay	0.83 – 34.7 ng g^{-1} dw	(Kwan, Takada, Mizukawa, Saha, et al., 2013)
	3	PBDEs in leachates from municipal solid waste dumping sites in tropical Asian countries: phase distribution and debromination	46 PBDEs	Leachates from solid waste dumping sites	10.5 – 19.2 ng L^{-1}	(Kwan, Takada, Mizukawa, Torii, et al., 2013)

Malaysia	1	Trace determination of waterborne organic pollutants in Klang River Basin	PCBs (equivalent of commerical mixtures Arochlor 1242, 1254 and 1260)	River water	n.d. – 4190 ng L^{-1}	(Tan et al., 1991)
	2	Geographical distribution of Persistent Organochlorines in air, water and sediments from Asia and Oceania, and their implications for global redistribution from lower latitudes	PCBs	Water from rural area	0.45 ng L^{-1}	(Iwata et al., 1994)
	3	Organochlorine contamination in fish and mussels from Cambodia and other Asian countries	PCBs (concentrations of individually resolved peak areas relative to an equivalent mixture standard of Aroclor (1016:1242:1254:1260; 1:1:1:1)	Green mussels *(Perna viridis)*	0.1 – 5.1 ng g^{-1} ww	(Monirith et al., 2000)
	4	Asia-Pacific mussel watch: monitoring contamination of persistent organochlorine compounds in coastal waters of Asian countries	PCBs (concentrations of individually resolved peak areas relative to an equivalent mixture standard of Aroclor (1016:1242:1254:1260; 1:1:1:1)	Green mussels *(Perna viridis)*	< 4.2–250 ng g^{-1} lw	(Monirith et al., 2003)
	5	Perfluorinated substances in tap water of Japan and	PFOA	Tap water	< LOD – 0.1 ng L^{-1}	(Lien et al., 2006)
				Surface water	< LOQ – 3.2	

		several countries and their relationship to surface water contamination			ng L^{-1}	
			PFOS	Tap water	< LOD – 0.1 ng L^{-1}	
				Suface water	< LOQ – 3.4 ng L^{-1}	
	6	Asian Mussel Watch Program: Contamination Status of Polybrominated Diphenyl Ethers and Organochlorines in Coastal Waters of Asian Countries	62 PCBs	Mussels	25 - 160 ng g^{-1} lw	(Ramu et al., 2007)
			14 PBDEs	Mussels	0.83 – 15.81 ng g^{-1} lw	
	7	Chemical contaminantion of Green Turtle (*Chelonia mydas*) Eggs in Peninsular Malaysia: Implications for Conservation and Public Health	83 PCBs	Green turtle (*Chelonia mydas*) eggs	0.15 – 3.69 ng g^{-1} ww	(van de Merwe et al., 2009)
			19 PBDEs		< LOD – 0.35 ng g^{-1} ww	
	8	Levels, Temporal Trends, and Tissue Distribution of Perfluorinated Surfactants in Freshwater Fish from Asian Countries	10 PFAS	Liver samples from freshwater fish	2.7 – 6.5 ng g^{-1} ww	(Murakami et al., 2011)
				Muscle samples from freshwater fish	0.3 – 0.5 ng g^{-1} ww	
	9	Perfluorooctanoic Acid (PFOA) and Perfluorooctane	PFOA	Surface river water	200 – 5940 ng L^{-1}	(Zainuddin et al., 2012)
			PFOS		710 – 43500	

		Sulfonate (PFOS) in surface water from the Langat River, Peninsular Malaysia			ng L^{-1}	
	10	Sedimentary PBDEs in urban areas of tropical Asian countries	23 PBDEs	Sediment in urban canals, rivers and marine bay	124 – 527 ng g^{-1} dw	(Kwan, Takada, Mizukawa, Saha, et al., 2013)
	11	PBDEs in leachates from municipal solid waste dumping sites in tropical Asian countries: phase distribution and debromination	46 PBDEs	Leachates from solid waste dumping sites	147 - 308 ng L^{-1}	(Kwan, Takada, Mizukawa, Torii, et al., 2013)
	12	Occurrence, distribution, and dichlorination of polychlorinated biphenyls and health risk assessment in Selangor River basin	44 PCBs	River water	0.13 – 7.67 ng L^{-1}	(Sakai et al., 2016)
	13	Examination of barnacles' potential to be used as bioindicators of persistent organic pollutants in coastal ecosystem: A Malaysia case study	62 PCBs	Barnacles (*Amphibalanus sp.*; *Tetraclita sp.*)	7.9 – 300 ng g^{-1} lw	(Vaezzadeh et al., 2021)
				Green mussel	14 – 180 ng g^{-1} lw	
			44 PBDEs	Barnacles (*Amphibalanus*	1.1 – 150 ng g^{-1} lw	

				sp.; *Tetraclita sp.*)		
				Green mussel	11 – 110 ng g^{-1} lw	
	14	Insights into the per- and polyfluoroalkyl substances-contaminated paper mill processing discharge: Detection, phytotoxicity, bioaccumulative profiling, and health risk verification	20 PFAS	Paper mill procesing discharge	2712.72 ng L^{-1} (accumulative concentration)	(Chow & Foo, 2023)
The Philippines	1	Isomer-Specific Accumulation and Toxic Assesment of Polychlorinated Biphenyls, including Coplanar congeners, in Cetaceans from the North Pacific and Asian coastal waters	Non-ortho PCBs	Blubbers of Cetaceans	PCB-77: 0.25 ng g^{-1} ww	(T. B. Minh et al., 2000)
					PCB-126: 0.15 ng g^{-1} ww	
					PCB-169: 0.35 ng g^{-1} ww	
			Mono-ortho PCBs		PCB-105: 25 ng g^{-1} ww	
					PCB-118: 80 ng g^{-1} ww	
					PCB-156: 12 ng g^{-1} ww	
	2	Recent contamination of Persistent chlorinated endocrine disrupters in	PCBs	Blubbers of Spinner dolphin and	2500 – 6200 ng g^{-1} ww	(T. Minh et al., 2000)

		Ceataceans from the North Pacific and Asian coastal waters		Fraser's dolphin		
	3	Organochlorine contamination in fish and mussels from Cambodia and other Asian countries	PCBs (concentrations of individually resolved peak areas relative to an equivalent mixture standard of Aroclor (1016:1242:1254:1260; 1:1:1:1)	Green mussels *(Perna viridis)*	0.69 - 36 ng g^{-1} ww	(Monirith et al., 2000)
	4	Asia-Pacific mussel watch: monitoring contamination of persistent organochlorine compounds in coastal waters of Asian countries	PCBs (concentrations of individually resolved peak areas relative to an equivalent mixture standard of Aroclor (1016:1242:1254:1260; 1:1:1:1)	Green mussels *(Perna viridis)*	22–640 ng g^{-1} lw	(Monirith et al., 2003)
	5	Global pollution monitoring of PCBs and Organochlorine Pesticides using Skipjack Tuna as a Bioindicator	117 PCB detected	Liver of Skipjack Tuna (*Katsuwonus pelamis)*	140 – 260 ng g^{-1} lw	(Ueno et al., 2003)
	6	Global pollution monitoring of Polybrominated Diphenyl Ethers Using Skipjack Tuna as a Bioindicator	11 PBDEs	Muscle tissue (pooled samples) of Skipjack Tuna (*Katsuwonus pelamis)*	14 ng g^{-1} lw	(Ueno et al., 2004)
	7	Asian Mussel Watch Program:	62 PCBs	Mussels	1100 - 1900 ng g^{-1} lw	(Ramu et al., 2007)

		Contamination Status of Polybrominated Diphenyl Ethers and Organochlorines in Coastal Waters of Asian Countries	14 PBDEs	Mussels	68.9 – 138.8 ng g^{-1} lw	
	8	Organic contaminants in the marine environment of Manila Bay, Philippines	13 PCBs	Sediment	0.1 – 1.47 ng g^{-1} dw	(Carvalho et al., 2009)
	9	PCBs in sediments and oysters of Manila Bay, the Philippines		Oysters (*Cassostrea gigas*)	6.7 – 72.9 ng g^{-1} dw	(Villeneuve et al., 2010)
	10	Polychlorinated Biphenyls in selected sites in Pasig River and Laguna Lake in the Philippines before and after a big flood event investigated under the UNU East Asia Regional POPs Monitoring Project	16 PCBs	River water	Before storm: 0.9 – 12.2 ng L^{-1} After storm: 6.1 – 32 ng L^{-1}	(Santiago & Rivas, 2012)
				Lake water	Before storm: 0.1 – 0.9 ng L^{-1} After storm: 2.9 – 10.8 ng L^{-1}	
	12	Sedimentary PBDEs in urban areas of tropical Asian countries	23 PBDEs	Sediment in urban canals, rivers and marine bay	0.56 – 3140 ng g^{-1} dw	(Kwan, Takada, Mizukawa, Saha, et al., 2013)

	13	PBDEs in leachates from municipal solid waste dumping sites in tropical Asian countries: phase distribution and debromination	46 PBDEs	Leachates from solid waste dumping sites	3.7 - 162 ng L^{-1}	(Kwan, Takada, Mizukawa, Torii, et al., 2013)
	14	Historical occurrences of polybrominated diphenyl ethers and polychlorinated biphenyls in Manila Bay, Philippines, and in the upper Gulf of Thailand	38 PCBs	Surface sediment	4.96 – 9.2 ng g^{-1} dw	(Kwan et al., 2014)
			36 PBDEs		0.67 – 5.12 ng g^{-1} dw	
	15	Prevalence of per- and polyfuoroalkyl substances (PFASs) in drinking and source water from two Asian countries	11 PFAS detected	Drinking water	9.08 – 11.6 ng L^{-1}	(Guardian et al., 2020)
			15 PFAS detected	Source water	15.5 – 35.7 ng L^{-1}	
	1	Persistent organic pollutants in Singapore's coastal marine environment: Part I, seawater	8 PCBs	Coastal water	0.22 – 61.76 ng L^{-1}	(Basheer et al., 2003)
	2	Occurrence of Polychlorinated Biphenyls and Polybrominated Diphenyl Ethers in green mussels (*Perna*	41 PCBs	Green mussels (*Perna viridis*)	6.1 – 82 ng g^{-1} dw	(Bayen et al., 2003)
			21 PBDEs		2.0 – 38 ng g^{-1} dw	

		viridis) from Singapore Southeast Asia				
	3	Asia-Pacific mussel watch: monitoring contamination of persistent organochlorine compounds in coastal waters of Asian countries	PCBs (concentrations of individually resolved peak areas relative to an equivalent mixture standard of Aroclor (1016:1242:1254:1260; 1:1:1:1)	Green mussels *(Perna viridis)*	90 ng g^{-1} lw	(Monirith et al., 2003)
	4	Analysis of persistent organic pollutants in marine sediments using a novel microwave assisted solvent extraction and liquid-phase microextraction technique	8 PCBs	Coastal sediment	10 – 61 ng g^{-1} dw	(Basheer et al., 2005)
	5	Persistent organic pollutants in mangrove food webs in Singapore	40 PCBs	Subsurface coastal water	0.17 – 7.1 ng L^{-1}	(Bayen et al., 2005)
				Mangrove sediment	0.59 – 1.86 ng g^{-1} dw	
				Organisms in mangrove food webs	0.6 – 190 ng g^{-1} ww	
			5 PBDEs	Subsurface coastal water	< LOD	
				Mangrove sediment	< LOD	

				Organisms in mangrove food webs	< LOD – 9.9 ng g^{-1} ww	
	6	Organochlorine pesticides, polychlorinated biphenyls and polybrominated diphenyl ethers in Singapore's coastal marine sediments	36 PCBs	Marine sediment	1.4 – 329.6 ng g^{-1} ww	(Wurl & Obbard, 2005b)
			Only BDE-47 detected		3.4 – 13.8 ng g^{-1} ww	
	7	Chlorinated pesticides and PCBs in the sea-surface microlayer and seawater samples of Singapore	36 PCBS	Sea water	0.05 – 1.8 ng L^{-1}	(Wurl & Obbard, 2005a)
	8	Distribution of organochlorine compounds in the sea-surface microlayer, water column and sediment of Singapore's coastal environment	37 PCBs	Sea water	0.06 – 6.98 ng L^{-1}	(Wurl & Obbard, 2006)
	9	Exposure and response of aquacultured oysters, *Crassostrea gigas*, to marine contaminants	40 PCBs	Oyster	Exposure experiment	(Bayen et al., 2007)
	10	Occurrence and source characterization of perfluorochemicals in an urban watershed	13 PFAS	Water from rivers and canals (dry season)	25.6 – 138.1 ng L^{-1}	(V. T. Nguyen et al., 2011)

				Water from rivers and canals (rainy season)	41.8 – 79.1 ng L^{-1}	
				Rainwater	5.2 – 11.5 ng L^{-1}	
	11	Occurrence, fate, and uxes of perfluorochemicals (PFCs) in an urban catchment: Marina Reservoir, Singapore	15 PFAS	Strom water for canals	64 – 92 ng L^{-1} (average total concentration)	(V. T. Nguyen et al., 2012)
			11 PFAS detected	Surface sediment from Marina watershed	3.5 – 6.3 ng g^{-1} dw	
	12	Co-extraction and simultaneous determination of multi-class hydro-phobic organic contaminants in marine sediments and biota using GC-EI-MS/MS and LC-ESI-MS/MS	28 PCBs	Mangrove sediment	0.68 ng g^{-1} dw (average value)	(Zhang et al., 2015a)
				Offshore sediment	0.077 ng g^{-1} dw (average value)	
				Marine organisms	0.45 – 1.31 ng g^{-1} ww	
			8 PBDEs	Mangrove sediment	0.031 ng g^{-1} dw (average value)	
				Offshore sediment	0.014 ng g^{-1} dw (average value)	
				Marine organisms	0.022 – 0.177 ng g^{-1} ww	

	13	Multi-residue analysis of legacy POPs and emerging organic contaminants in Singapore's coastal waters using gas chromatography–triple quadrupole tandem mass spectrometry	28 PCBs	Coastal water	0.0092 – 0.151 ng L^{-1}	(Zhang et al., 2015b)
	14	Fate and transport of perfluoro- and polyfluoroalkyl substances including perfluorooctane sulfonamides in a managed urban water body	15 PFAS	Surface water (dissolved phase)	Rainy season: 174 ng L^{-1} Dry season: 138 ng L^{-1}	(T. V. Nguyen et al., 2016)
				Surface water (suspended phase)	Rainy season: 158 ng L^{-1} Dry season: 15 ng L^{-1}	
				Surface sediment	Rainy season: 14.3 ng g^{-1} dw Dry season: 15.3 ng g^{-1} dw	
	15	Characterization of occurrence, sources and sinks of perfluoroalkyl and polyfluoroalkyl substances (PFASs) in a tropical urban catchment	22 PFAS	Water from urban catchment	23 – 39 ng L^{-1} (average of median values)	(Chen et al., 2017)
				Surface sediment	March: 0.15 – 2.1 ng g^{-1} dw September: 2.64 – 8.75 ng g^{-1} dw	

	16	Occurrence and distribution of halogenated flame retardants in an urban watershed: Comparison to polychlorinated biphenyls and organochlorine pesticides	28 PCBs	Water from open water bodies	0.017 – 0.043 ng L^{-1}	(Wang & Kelly, 2017)
				Water from catchment canals	0.025 – 0.34 ng L^{-1}	
				Bottom Sediment	6.48 – 212.3 ng g^{-1} dw	
				Suspended particulate matters	9.86 – 27.92 ng g^{-1} dw	
			7 PBDEs	Water from open water bodies	0.005 – 0.16 ng L^{-1}	
				Water from catchment canals	0.017-0.2 ng L^{-1}	
				Bottom Sediment	0.75 – 6.14 ng g^{-1} dw	
				Suspended particulate matters	0.029 – 3.25 ng g^{-1} dw	
	17	Perfluoroalkyl and polyfluoroalkyl substances removal in a full-scale tropical constructed wetland system treating landfill leachate	18 PFAS	Raw landfill leachate	1269 – 7661 ng L^{-1}	(Yin et al., 2017)

	18	Assessing bioaccumulation behaviour of hydrophobic organic contaminants in a tropical urban catchment	28 PCBs	Plankton	164.9 – 4143.3 ng g^{-1} lw	(Wang & Kelly, 2018)
				Apple snails	79.8 – 2216.9 ng g^{-1} lw	
				Tilapia	180.9 – 8471.9 ng g^{-1} lw	
				Peacock bass	569.9 – 19223 ng g^{-1} lw	
			7 PBDEs	Plankton	26.51 – 935.16 ng g^{-1} lw	
				Apple snails	10.14 – 230.95 ng g^{-1} lw	
				Tilapia	30.87 – 2729.43 ng g^{-1} lw	
				Peacock bass	129.1 – 1744.53 ng g^{-1} lw	
	19	Sorption and bioaccumulation behavior of multi-class hydrophobic organic contaminants in a tropical marine food web	28 PCBs	Marine tropical species	0.14 – 0.58 ng g^{-1} ww (average values)	(Zhang & Kelly, 2018)
			7 PBDE		n.d. – 0.079 ng g^{-1} ww (average values)	
	20		18 PFAS	Surface water	30.6 – 47.8	(Chen et al., 2019)

		Multi-compartment distribution of perfluoroalkyl and polyfluoroalkyl substances (PFASs) in an urban catchment system			ng L^{-1}	
				Suspended particles	<26.8 – 1284 ng g^{-1} dw	
				Sediment	<2.84 – 8.38 ng g^{-1} dw	
Thailand	1	Geographical distribution of Persistent Organochlorines in air, water and sediments from Asia and Oceania, and their implications for global redistribution from lower latitudes	PCBs	Water from urban areas and industrial canals	0.24 – 4.4 ng L^{-1}	(Iwata et al., 1994)
				Sediment from river in urban zones and industrial canals	11 – 520 ng g^{-1} dw	
	2	Contaminant by Butylin compounds and Organochlorine residues in Green mussels (*Perna viridis*, L.) from Thailand coastal waters	PCBs (concentrations of individually resolved peak areas relative to an equivalent mixture standard of Aroclor (1016:1242:1254:1260)	Green mussels (*Perna viridis*)	< 0.01 - 20 ng g^{-1} ww	(Kan-Atireklap et al., 1997)
	3	Dioxins/Furans and Polychlorinated Biphenyls (PCBs) in Dugongs from the Thailand Coast	14 PCBs	Dugong (*Dugong dugon*)	3.6 – 4.7 ng g^{-1} fat weight	(Kumar et al., 2003)
				Seagrass collected in Dugong stomach	0.054 ng g^{-1} dw	
	4	Environmental occurrence of	PCBs	Green mussels (*Perna viridis*)	0.32 – 3.7 ng g^{-1} ww	(Cheevaporn et al., 2005)

		Organochlorines in the East Coast of Thailand		Oysters (*Crassostrea commercialis*)	1.32 – 10.29 ng g^{-1} ww	
	5	Perfluorinated substances in tap water of Japan and several countries and their relationship to surface water contamination	PFOA	Tap water	< LOD – 4.6 ng L^{-1}	(Lien et al., 2006)
				Surface water	< LOQ – 8.8 ng L^{-1}	
			PFOS	Tap water	0.1 – 1.9 ng L^{-1}	
				Suface water	< LOQ – 1.1 ng L^{-1}	
	6	Reconstruction of pollution history of organic contaminants in the upper Gulf of Thailand by using sediment cores: First report from Tropical Asia Core (TACO) project	24 PCBs	Marine sediment core (surface to 60 cm depth)	n.d. – 2 ng g^{-1} dw	(Boonyatumanond et al., 2007)
	7	PCBs contamination in seafood species at the Eastern Coast of Thailand	49 PCBs	Mussels	0.19 – 6.22 ng g^{-1} ww	(Jaikanlaya et al., 2009)
				Oysters	0.54 – 7.53 ng g^{-1} ww	
				Shrimps	0.05 – 2.7 ng g^{-1} ww	
	8	Contamination of perfluorinated compounds (PFCs) in Chao Phraya River and Bangpakong River, Thailand	8 PFAS	River water	1.2 – 9 ng L^{-1} (average values)	(Kunacheva et al., 2009)
				Industrial effluent	233 ng L^{-1} (average values)	

	9	Perfluorinated compounds contamination in tap water and bottled water in Bangkok, Thailand	5 PFAS	Raw water	25.8 ng L^{-1} (average values)	(Kunacheva et al., 2010)
				Tap water	6.03 ng L^{-1} (average values)	
				Bottled water	11.64 ng L^{-1} (average values)	
	10	Mass flows of perfluorinated compounds (PFCs) in central wastewater treatment plants of industrial zones in Thailand	10 PFAS	Influent in from wastewater treatment plants (WWTP)	673 - 847 ng L^{-1} (average values)	(Kunacheva, Tanaka, Fujii, Boontanon, Musirat, Wongwattana, et al., 2011)
				Effluent in from WWTP	661 – 1143 ng L^{-1} (average values)	
	11	Determination of perfluorinated compounds (PFCs) in solid and liquid phase river water samples in Chao Phraya River, Thailand	10 PFAS	River water – dissolved phase	4.3 – 24.2 ng L^{-1}	(Kunacheva, Tanaka, Fujii, Boontanon, Musirat, & Wongwattana, 2011)
				River water – solid phase	2.8 – 11 ng g^{-1} dw	
	12	Levels, Temporal Trends, and Tissue Distribution of Perfluorinated Surfactants in Freshwater Fish from Asian Countries	10 PFAS	Freshwater fish	n.d.	(Murakami et al., 2011)

	13	Assessment of persistent organic pollutants in sediments from Lower Mekong River Basin	62 PCBs	Mekong River sediment	0.18 - 47 ng g^{-1} dw	(Sudaryanto et al., 2011)
	14	Occurrence of Perfluorooctane Sulfonate in the Water Environment of Bangkok, Thailand	PFOS	River water	< LOQ – 20.05 ng L^{-1}	(Boontanon et al., 2013)
				Residential tap water	0.4 ng L^{-1} (average value)	
				Bottled water	0.5 ng L^{-1} (average value)	
			PFOA	River water	0.68 – 20.41 ng L^{-1}	
	15	Potential release of PCBs from plastic scientific gear to fringing coral reef sediments in the Gulf of Thailand	21 PCBs detected	Marine bottom sediment	2.16 – 3.27 ng g^{-1} dw (average values)	(G. H. Hong et al., 2013)
	16	Sedimentary PBDEs in urban areas of tropical Asian countries	23 PBDEs	Sediment in urban canals, rivers and marine bay	42.8 – 590 ng g^{-1} dw	(Kwan, Takada, Mizukawa, Saha, et al., 2013)
	17	PBDEs in leachates from municipal solid waste dumping sites in tropical Asian countries: phase distribution and debromination	46 PBDEs	Leachates from solid waste dumping sites	24.1 - 133000 ng L^{-1}	(Kwan, Takada, Mizukawa, Torii, et al., 2013)

	18	Historical occurrences of polybrominated diphenyl ethers and polychlorinated biphenyls in Manila Bay, Philippines, and in the upper Gulf of Thailand	38 PCBs	Surface sediment	1.79 ng g^{-1} dw	(Kwan et al., 2014)
			36 PBDEs		1.94 ng g^{-1} dw	
	19	Fate and risk Assessment of Perfluoroalkyl substances (PFASs) in water treatment plants and tap water in Bangkok, Thailand	11 PFAS	Influent – Effluent from conventional WWTP	2.39 – 1.88 ng L^{-1}	(Tabtong et al., 2015)
				Influent – Effluent from advanced WWTP	71.07 – 9.73 ng L^{-1}	
			6 PFAS	Tap water	1.07 – 1.15 ng L^{-1}	
				Drinking water	0.58 – 0.83 ng L^{-1}	
	20	Levels of perfluorinated compounds (PFCs) in groundwater around improper municipal and industrial waste disposal sites in Thailand and health risk assessment	6 PFAS detected	Groundwater from municipal waste disposal site	1.68 – 2.75 ng L^{-1}	(Hongkachok et al., 2017)
				Groundwater from industrial waste disposal site	2.64 – 42 ng L^{-1}	
	21	Prevalence of per- and polyfuoroalkyl	12 PFAS detected	Drinking water	7.16 – 59.5 ng L^{-1}	(Guardian et al., 2020)

		substances (PFASs) in drinking and source water from two Asian countries		Source water	22.2 – 65.6 Ng L^{-1}	
	22	Determination of perfluorooctane sulphonate and perfluorooctanoic acid in seafood and water from Map Ta Phut Industrial Estate area, Thailand	PFOA	Water from river and canals	Dry season: 1.3 – 59.3 ng L^{-1} Wet season: 3.8 – 31 ng L^{-1}	(Lertassavakorn et al., 2021)
				Marine orgamisms	Dry season: < LOD – 0.42 ng g^{-1} ww Wet season: < LOD – 0.28 ng g^{-1} ww	
			PFOS	Water from river and canals	Dry season: 7.4 – 113.9 ng L^{-1} Wet season: 4.5 – 465.7 ng L^{-1}	
				Marine orgamisms	Dry season: < LOD – 6.72 ng g^{-1} ww Wet season: < LOD – 3.82 ng g^{-1} ww	
	23	Per- and polyfluoroalkyl substances in groundwater in Thailand: occurrence,	6 PFAS detected	Groundwater	1.67 – 42.0 ng L^{-1}	(Hongkachok et al., 2023)

		source identification and spatial distribution				
Vietnam	1	Geographical distribution of Persistent Organochlorines in air, water and sediments from Asia and Oceania, and their implications for global redistribution from lower latitudes	PCBs	Sediment from rivers in urban areas; and from paddy fields and mangroves	0.57 – 8 ng L^{-1}	(Iwata et al., 1994)
				Sediment from rivers and canals in urban areas; and from paddy fields and mangroves	< LOD - 630 ng g^{-1} dw	
	2	Organochlorine pesticides and PCBs in the Red River Delta, North Vietnam	PCBs (relative to an equivalent mixture standard of Aroclor)	Freshwater sediment	Dry season: 5.64- 10.56 ng g^{-1} dw Rainy season: 2.17 – 3.83 ng g^{-1} dw	(Duch Nhan et al., 1998)
				Estuarine sediment	Dry season: 1.62 – 3.27 ng g^{-1} dw Rainy season: 0.97 – 1.35 ng g^{-1} dw	
				Aquatic species	Rainy season: 5.1 – 34 ng g^{-1} dw	
	3	Contamination by PCB's, DDT's and heavy metals in sediments of	6 PCBs	Sediment from canals	< LOD – 123.46 ng g^{-1} dw	(Phuong et al., 1998)

		Ho Chi Minh City's canals, Vietnam				
	4	Organochlorine pesticides and PCBs along the coast of north Vietnam	PCBs (Aroclor 1254 equivalent)	Coastal sediment	0.47 – 28.1 ng g^{-1} dw	(Duc Nhan et al., 1999)
				Clams (*Meretrix meretrix*)	5.1 – 25.3 ng g^{-1} dw	
	5	Organochlorine contamination in fish and mussels from Cambodia and other Asian countries	PCBs (concentrations of individually resolved peak areas relative to an equivalent mixture standard of Aroclor (1016:1242:1254:1260; 1:1:1:1)	Green mussels *(Perna viridis)*	0.2 – 3.4 ng g^{-1} ww	(Monirith et al., 2000)
	6	Chlorinated pesticides and PCBs in sediments and molluscs from freshwater canals in the Hanoi region	13 PCBs	Sediment	0.73 – 33.68 ng g^{-1} ww	(Duc Nhan et al., 2001)
				Molluscs	5.45 – 58.12 ng g^{-1} ww	
	7	Asia-Pacific mussel watch: monitoring contamination of persistent organochlorine compounds in coastal waters of Asian countries	PCBs (concentrations of individually resolved peak areas relative to an equivalent mixture standard of Aroclor (1016:1242:1254:1260; 1:1:1:1)	Green mussels *(Perna viridis)*	21–450 ng g^{-1} lw	(Monirith et al., 2003)
	8	Perfluorinated substances in tap water of Japan and several countries and	PFOA	Tap water	< LOD	(Lien et al., 2006)
				Surface water	< LOQ – 6.6 ng L^{-1}	
			PFOS	Tap water	< LOD	

		their relationship to surface water contamination		Suface water	< LOQ – 1.3 ng L^{-1}	
	9	Contamination by Polybrominateed Diphenyl Ethers and Persistent Organochlorines in catfish and feed from Mekong River Delta, Vietnam	62 PCBs	Common catfish	0.91 – 27 ng g^{-1} lw	(Hung Minh et al., 2006)
				Catfish near dumpsite	37 – 77 ng g^{-1} lw	
			6 PBDE	Common catfish	0.12 – 1.4 ng g^{-1} lw	
				Catfish near dumpsite	1.3 – 3.7 ng g^{-1} lw	
	10	Polychlorinated biphenyls in sediments of the Tam Giang-Cau Hai Lagoon, Central Vietnam	53 PCBs	Surface sediments from lagoon	10.2 – 24.5 ng g^{-1} dw	(Frignani et al., 2007)
	11	Distribution of Persistent Organic Pollutants and Polycyclic Aromatic Hydrocarbons in sediment samples from Vietnam	PCBs	Sediment	0.11 – 105.91 ng g^{-1} dw	(Kishida et al., 2007)
	12	Persistent Organic Pollutants in Sediments from Sai Gon–Dong Nai River Basin, Vietnam: Levels and Temporal Trends	62 PCBs	Sediment from river and canal	0.33 – 150 ng g^{-1} dw	(N. H. Minh, Minh, Iwata, et al., 2007)
				Estuarine sediment	0.49 – 2.4 ng g^{-1} dw	
	13	Pollution sources and ccurrences of selected persistent organic	PCBs	Freshwater sediment	Dry season: 0.12 – 3.7 ng g^{-1} dw	(N. H. Minh, Minh, Kajiwara, et al., 2007)

		pollutants (POPs) in sediments of the Mekong River delta, South Vietnam			Rainy season: 0.04 – 9.2 ng g^{-1} dw	
	14	Asian Mussel Watch Program: Contamination Status of Polybrominated Diphenyl Ethers and Organochlorines in Coastal Waters of Asian Countries	62 PCBs	Mussels	26 – 290 ng g^{-1} lw	(Ramu et al., 2007)
			14 PBDEs	Mussels	0.66 – 5.5 ng g^{-1} lw	
	15	Agrochemical and polychlorobyphenyl (PCB) residues in the Mekong River delta, Vietnam	13 PCBs	Water	n.d.	(Carvalho et al., 2008)
				Sediment	0.11 – 2.02 ng g^{-1} dw	
				Molluscs	1.89 – 19.37 ng g^{-1} dw	
	16	Persistent organochlorine residues in estuarine and marine sediments from Ha Long Bay, Hai Phong Bay, and Ba Lat Estuary, Vietnam	23 PCBs	Sediment	0.04 – 18.7 ng g^{-1} dw	(S. H. Hong et al., 2008)
	17	Analysis of biomagnification of persistent organic pollutants in the aquatic food web of the Mekong Delta, South Vietnam using	60 PCBs	Biota	0.017 – 8.9 ng g^{-1} ww	(Ikemoto et al., 2008)

		stable carbon and nitrogen isotopes				
	18	Polychlorinated Biphenyl Congeners in the Aquatic Environment of the Mekong River, South of Vietnam	13 PCBs	Sediment	0.11 – 2.02 ng g^{-1} dw	(Carvalho et al., 2009)
				Clams	1.89 – 19.37 ng g^{-1} dw	
	19	Characteristics of the abundance of polychlorinated dibenzo-p-dioxin and dibenzofurans, and dioxin-like polychlorinated biphenyls in sediment samples from selected Asian regions in Can Gio, Southern Vietnam and Osaka, Japan	12 PCBs	Sediment from mangrove forest and lagoon	0.25 – 0.26 ng g^{-1} dw (average values)	(Kishida et al., 2010)
				Sediment from urban lakes	12.7 ng g^{-1} dw (average values)	
	20	Recent levels of organochlorine pesticides and polychlorinated biphenyls in sediments of the sewer system in Hanoi, Vietnam	7 PCBs	Sediment from urban and river lakes	0.33 – 96 ng g^{-1} dw	(Hoai et al., 2010)
	21	PCBs in Central Vietnam coastal lagoons: Levels and trends	101 PCBs	Surficial sediment	1.32 – 24.5 ng g^{-1} dw	(Giuliani et al., 2011)

		in dynamic environments				
	22	Levels, Temporal Trends, and Tissue Distribution of Perfluorinated Surfactants in Freshwater Fish from Asian Countries	10 PFAS	Liver samples from freshwater fish	11.7 – 43 ng g^{-1} ww	(Murakami et al., 2011)
				Muscle samples from freshwater fish	0.1 – 0.5 ng g^{-1} ww	
	23	Contamination by perfluorinated compounds in water near waste recycling and disposal sites in Vietnam	17 PFAS	Water	n.d. – 170 ng L^{-1}	(Kim et al., 2013)
	24	Sedimentary PBDEs in urban areas of tropical Asian countries	23 PBDEs	Sediment in urban canals, rivers and marine bay	3.01 – 166 ng g^{-1} dw	(Kwan, Takada, Mizukawa, Saha, et al., 2013)
	25	PBDEs in leachates from municipal solid waste dumping sites in tropical Asian countries: phase distribution and debromination	46 PBDEs	Leachates from solid waste dumping sites	38.6 – 48.1 ng L^{-1}	(Kwan, Takada, Mizukawa, Torii, et al., 2013)
	26	PBDEs and PCBs in sediments of the Thi Nai Lagoon (Central Vietnam) and soils from its mainland	127 PCBs	Sediment from lagoon	0.47 – 6.4 ng g^{-1} dw	(Romano et al., 2013)
			9 PBDEs detected		< LOD – 9.62 ng g^{-1} dw	

	27	Screening and analysis of 940 organic micro-pollutants in river sediments in Vietnam using an automated identification and quantification database system for GC–MS	24 PCBs	River sediment	n.d. – 43.1 ng g^{-1} dw	(H. T. Duong et al., 2014)
	28	Residue of Dioxin-Like Polychlorinated Biphenyls (DL-PCBs) in sediment from CauBay River, Vietnam	12 PCBs	River sediment	9.4 – 17.4 ng g^{-1} dw	(Toan, 2014)
	29	Occurrence of perfluoroalkyl acids in environmental waters in Vietnam	10 PFAS detected	Water from rivers and canals	0.11 – 46.03 ng L^{-1}	(H. T. Duong et al., 2015)
	30	Distribution, accumulation profile, and risk assessment of polybrominated diphenyl ethers in sediment from lake and river systems in Hanoi Metropolitan Area, Vietnam	7 PBDEs	Sediment from urban lakes and rivers	0.03 – 17.5 ng g^{-1} dw	(Mai et al., 2015)
	31	Flame retardant emission from e-waste recycling operation in northern Vietnam: Environmental occurrence of	14 PBDEs	Sediment from river near e-waste recycling sites	0.43 – 350 ng g^{-1} dw	(Matsukami et al., 2015)

		emerging organophosphorus esters used as alternatives for PBDE				
	32	Residues of Polychlorinated Biphenyls (PCBs) in Sediment from CauBay River and Their Impacts on Agricultural Soil, Human Health Risk in KieuKy Area, Vietnam	84 PCBs	River sediment	2.3 – 4.2 ng g^{-1} dw	(Toan & Quy, 2015)
	33	Characteristics of marine environmental geochemistry from Ha Tinh to Quang Nam (60-100m water depth)	6 PCBs	Marine sediment	0.32 – 5.05 ng g^{-1} dw	(Huy et al., 2016)
	34	Distribution and depth profiles of polychlorinated dibenzo-p-dioxins, polychlorinated dibenzofurans, and polychlorinated biphenyls in sediment collected from offshore waters of Central Vietnam	8 PCBs	Marine sediment	0.036 – 0.158 ng g^{-1} dw	(Tri et al., 2016)

	35	Polybrominated diphenyl ethers in plastic products, indoor dust, sediment and fish from informal e-waste recycling sites in Vietnam: a comprehensive assessment of contamination, accumulation pattern, emissions, and human exposure	8 PBDEs	Sediment from drainage canals	1.31 – 1710 ng g^{-1} dw	(Anh et al., 2017)
				Fish nearby e-waste recycling sites	21.6 – 1380 ng g^{-1} lw	
	36	A nationwide survey of perfluorinated alkyl substances in waters, sediment and biota collected from aquatic environment in Vietnam: Distributions and bioconcentration profiles	13 PFAS	Surface water	< LOQ – 14.6 ng L^{-1}	(Lam et al., 2017)
			8 PFAS detected	Surficial sediment	< LOQ – 23.4 ng L^{-1}	
			13 PFAS	Biota	< LOQ – 26.2 ng g^{-1}, ww	
	37	Concentrations of polybrominated diphenyl ethers and alternative flame retardants in surface soils and river sediments from an electronic waste-processing area in	13 PBDEs	River sediment	**2012**: 2.78 -353.7 ng g^{-1} dw **2013**: 7.5 – 1070.4. ng g^{-1} dw **2014**: 36.4 – 3780.9 ng g^{-1} dw	(Matsukami et al., 2017)

		northern Vietnam, 2012-2014				
	38	Where has the pollution gone? A survey of organic contaminants in Ho Chi Minh city / Saigon River (Vietnam) bed sediments	7 PCBs	Sediment from river and urban canals	< LOD – 18.73 ng g^{-1} dw	(Babut et al., 2019)
			PBDEs		< LOD	
			6 PFAS detected		< LOD – 1.18 ng g^{-1} dw	
	39	When research meets NGOs: The GVC-UCODEP project in the Bắc Giang Province and Cầu River (Northern Vietnam) and its feedback on national monitoring programs	127 PCBs detected	Sediment from rivers, canals and ponds	2.9 – 31 ng g^{-1} dw	(Giuliani et al., 2019)
			14 PBDEs		0.73 – 57 ng g^{-1} dw	
	40	The interactive effect of the season and estuary position on the concentration of persistent organic pollutants in water and sediment from the Cua Dai estuary in Vietnam	7 PCBs	Estuarine water	30 – 1050 ng L^{-1}	(T. X. Nguyen et al., 2019)
				Estruarine sediement	24.7 – 329.7 ng g^{-1} dw	
	41	Endocrine Disrupting Compounds (EDCs) in Surface Waters of the KimNguu River, Vietnam	6 PCBs	Water from urban river	Dry season: 2.33 – 8.93 ng L^{-1} Rainy season: 0.95 – 4.19 ng L^{-1}	(Quynh & Toan, 2019)

			7 PBDEs		Dry season: 2.19 – 7.08 ng L^{-1} Rainy season: 0.96 – 3.17 ng L^{-1}	
	42	Distributions and seasonal variations of organochlorine pesticides, polychlorinated biphenyls, and polybrominated diphenyl ethers in surface sediment from coastal areas of central Vietnam	7 PCBs	Coastal surface sediment	9.72 - 3730 ng g^{-1} dw	(Tham et al., 2019)
			8 PBDEs		11.8 – 311 ng g^{-1} dw	
	43	Occurrence of organochlorine pesticides and polychlorinated biphenyls in fish in Cau Hai lagoon of Central Vietnam: Human health risk sediment and assessment	7 PCBs	Surface sediment	n.d.	(T. A. M. Tran et al., 2019)
				Fish from coastal lagoon	25.9 – 43 ng g^{-1} lw	
	44	Contamination status and temporal trends of persistent toxic substances in sediment cores from coastal	7 PCBs	Sediment cores	6.7 – 91 ng g^{-1} dw	(Tham et al., 2020)
			8 PBDEs		13 – 280 ng g^{-1} lw	

		areas of central Vietnam				
	45	Endocrine disrupting compounds in sediment from KimNguu river, Northern area of Vietnam: a comprehensive assessment of seasonal variation, accumulation pattern and ecological risk	18 PCBs	Sediment	Dry season: 5.75 – 36.34 ng g^{-1} dw Rainy season: 4.73 – 30.43 ng g^{-1} dw	(Toan et al., 2020)
			7 PBDEs		Dry season: 5.62 – 12.49 ng g^{-1} dw Rainy season: 4.37 – 10.68 ng g^{-1} dw	
	46	Comprehensive monitoring of more than 1000 Organic micro-pollutants in drainage water: Case study in a rural village with end-of-life vehicle processing activities in Northern Vietnam	64 PCBs detected	Water from drainage canal	3.3 – 5 ng L^{-1}	(Anh et al., 2021)
			BDE-28, -47, -209	Drainage water and water from nearby pond	1.87 – 2.99 ng L^{-1}	
	47	Comprehensive determination of polychlorinated biphenyls and brominated flame retardants in surface sediment samples from Hanoi urban area, Vietnam: Contamination status,	209 PCBs	Sediment	Dry season: 1.7 – 48.8 ng g^{-1} dw Rainy season: 2.3 - 45 ng g^{-1} dw	(Hoang et al., 2021)
			41 PBDEs		Dry season: 0.2 – 37.2 ng g^{-1} dw	

		accumulation profiles, and potential ecological risks			Rainy season: 0.69 – 61.4 ng g^{-1} dw	
	48	Occurrence of perfluorinated chemicals (PFCs) in leachate, surface water and sediment collected from Tay Mo landfill and surrounding areas	16 PFAS	Leachate	103 – 328 ng L^{-1}	(Ngoc et al., 2021)
				Urban river and pond	3.81 – 22.4 ng L^{-1}	
				Sediment from land field	3.52 – 6.7 ng g^{-1} dw	
				Sediment from urban river and pond	1.57 and 1.34 ng g^{-1} dw, respectively (average values)	
	49	Distribution of perfluoroalkyl substances (PFASs) in the water of the Bac Hung Hai River, Van Giang district, Hung Yen province, Vietnam	10 PFAS	River water	1 – 38.8 ng L^{-1}	(Tuan Do et al., 2021)
	50	Determination of Per- and Polyfluoroalkyl substances in craft villages and industrial environments of Vietnam	8 PFAS	Wastewater	< LOD – 59.5 ng L^{-1}	(Phung et al., 2021)
	51	What's in the water? – Target and suspect screening of contaminants	PFHxA	Raw water	0.91 ng L^{-1}	(Tröger et al., 2021)
			PFHpA		0.45 ng L^{-1}	
			PFOA		1.08 ng L^{-1}	
			PFNA		0.16 ng L^{-1}	

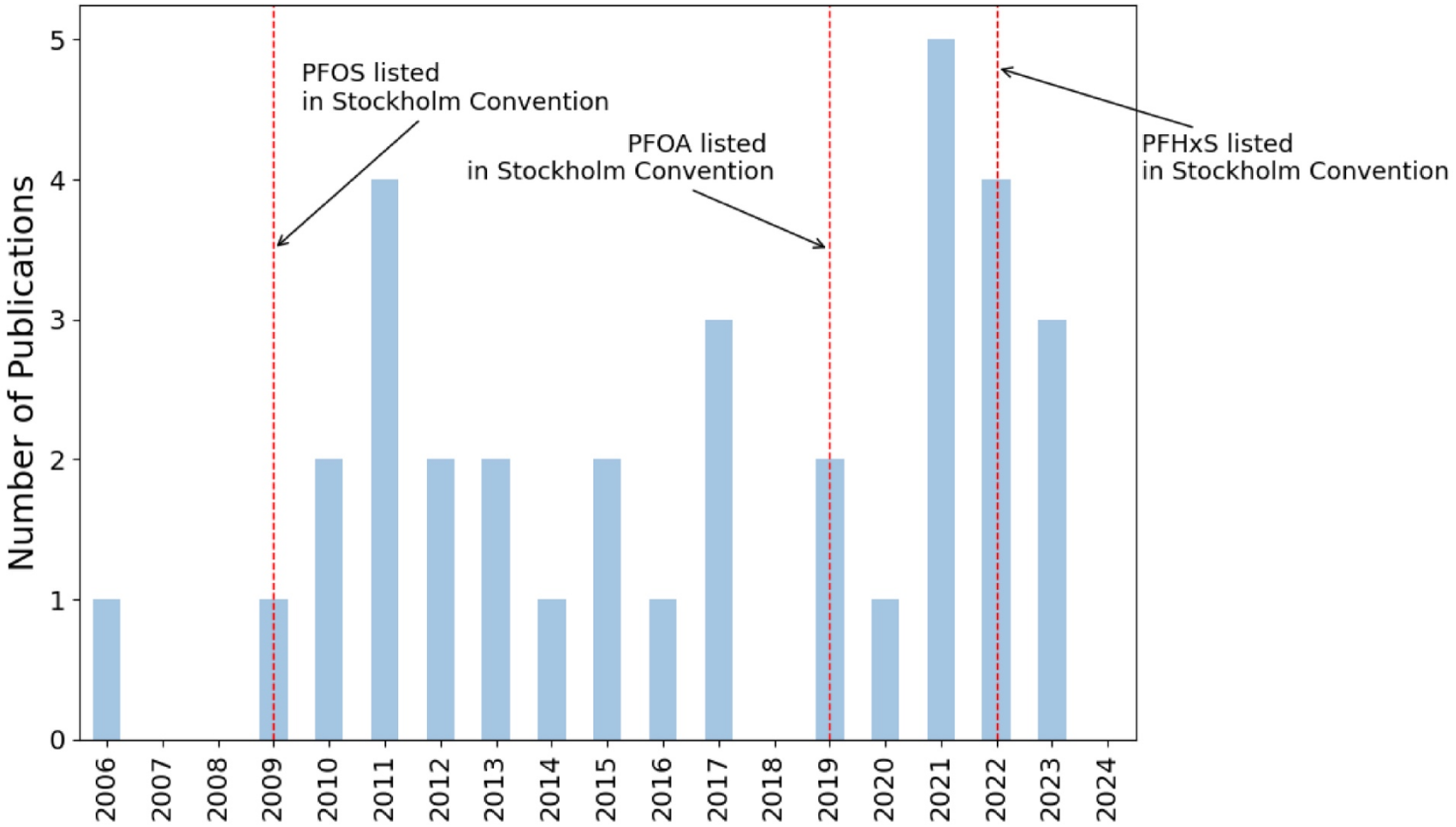
		of emerging concern in raw water and drinking water from Europe and Asia	PFHpA	Drinking water	0.26 ng L^{-1}	
			PFOA		0.42 ng L^{-1}	
			PFNA		0.13 ng L^{-1}	
	52	Marine sediment quality at coastal monitoring stations in Vietnam 2016–2020	6 PCBs	Marine sediment	0.22 – 60.96 ng g^{-1} dw	(T. N. Duong et al., 2022)
	53	Perfluoroalkyl substances (PFAS) in freshwater fish from urban lakes in Hanoi, Vietnam: concentrations, tissue distribution, and implication for risk assessment	17 PFAS	Fish blood	5.2 – 29 ng mL^{-1}	(Hoa et al., 2022)
				Fish liver	2.7 – 6.6 ng g^{-1} ww	
				Fish muscle	0.51 – 2.6 ng g^{-1} ww	
	54	Contamination of perfluorooctane sulfonic acid (PFOS) and perfluorooctanoic acid (PFOA) in sediment of the Cau River, Vietnam	PFOA	Surface river sediment	0.17 – 1.78 ng L^{-1}	(H. Le Tran et al., 2022)
			PFOS		1.19 – 4.73 ng L^{-1}	
	55	Accumulation and distribution of persistent organic pollutants in surface sediment of the estuarine region and correlation between genetic and	7 PCBs	Estuarine sediment	1.52 – 19.53 ng g^{-1} dw	(Le et al., 2022)

		environmental factors: A case study				
	56	Perfluoroalkyl substances in freshwater and marine fish from northern Vietnam: Accumulation levels, profiles, and implications for human consumption	9 PFAS	Freshwater fish	0.08 – 8.06 ng g^{-1} ww	(Vi et al., 2022)
				Marine fish	0.22 – 3.62 ng g^{-1} ww	
	57	Sediment contamination with polybrominated diphenyl ethers and alternative brominated flame retardants: case study in urban lakes of Hanoi, Vietnam	41 PBDEs	Sediment from urban lakes	1.1 – 25.71 ng g^{-1} dw	(Hoang et al., 2023)
	58	Occurrence, distribution, and risk assessment of halogenated organic pollutants (HOPs) in marine fish muscle: The case study of Vietnam	PCBs	Marine fish	4.5 – 711.6 ng g^{-1} lw	(Tran-Lam et al., 2023)
	59	Analysis of perfluoroalkyl compounds in marine fish muscle samples by liquid chromatography	23 PFAS	Marine fish	9 -28.6 ng g^{-1} ww	(Vu-Duc et al., 2023)

		tandem mass spectrometry				
	60	Halogenated organic pollutants (HOPs) in marine fish collected from central-northern coastal waters in Vietnam	28 PCBs	Marine fish	< MLD – 822 ng g^{-1} lw	(Duc et al., 2024)
			8 PBDEs		< MLD – 320 ng g^{-1} lw	
	61	Evaluation of chemical exposure to the sediment and green mussels (*Perna viridis*) at some coastal sites in Northern Vietnam	5 PBDEs	Coastal sediment	0.06 – 7.26 ng g^{-1} dw	(D. T. Nguyen et al., 2024)
				Green mussels (*Perna viridis*)	0.06 – 23.5 ng g^{-1} ww	

LOD: limit of detection; LOQ: limit of quantification; MLD: method limit of detection; n.d.: not detected; ww: wet weight; lw: lipid weight; dw: dry weight

Annex 7: Number of publications dealing with PFAS occurrence in the aquatic environment of the Southeast Asian region from 2006 (first reported occurrence) to 2024.



Annex

Annex 8: Coordinates, total organic carbon (TOC %), size distribution of sediment samples from 21 sampling stations.

Sampling stations	Latitude	Longitude	November 2022				March 2023				June 2023				September 2023			
			TOC (%)	Sand (%)	Silt (%)	Clay (%)	TOC (%)	Sand (%)	Silt (%)	Clay (%)	TOC (%)	Sand (%)	Silt (%)	Clay (%)	TOC (%)	Sand (%)	Silt (%)	Clay (%)
R00	21.1136	105.7231	0.54	25.77	73.96	0.13	0.34	59.57	40.43	0.00	0.78	43.35	56.55	0.09	1.05	29.84	70.06	0.09
R01	21.0932	105.7713	1.31	40.61	59.31	0.07	0.32	57.24	42.76	0.00	1.22	33.75	66.25	0.00	1.02	48.04	51.96	0.00
R02	21.0572	105.8553	0.34	55.75	44.27	0.00	0.66	30.77	69.16	0.07	0.63	37.39	62.61	0.00	1.16	39.05	60.87	0.08
R03	21.0028	105.8846	1.15	23.69	75.99	0.31	0.64	31.70	68.20	0.1	0.86	61.95	37.98	0.06	0.74	25.95	73.95	0.10
R04	20.9469	105.8840	0.35	80.75	19.25	0.00	0.52	95.03	4.97	0.00	0.88	63.72	36.28	0.00	0.19	76.48	23.53	0.00
R05	20.9309	105.8816	0.70	35.27	64.74	0.00	0.86	43.16	56.84	0.00	0.66	32.55	67.38	0.06	0.78	40.69	59.31	0.00
R06	20.8802	105.9191	0.72	49.03	50.95	0.00	0.33	53.81	46.18	0.00	0.65	38.43	61.57	0.00	0.68	41.35	58.65	0.00
R07	20.8061	105.9145	0.42	48.04	51.95	0.00	1.06	46.60	53.32	0.08	0.95	36.09	63.84	0.07	0.73	40.23	59.70	0.07
R08	20.7459	105.9665	1.21	32.34	67.50	0.15	1.06	33.96	65.95	0.09	1.19	36.24	63.61	0.14	1.23	31.24	68.65	0.11
R09	20.6875	106.0288	0.64	38.57	61.33	0.10	0.78	36.37	63.54	0.08	0.46	65.13	34.83	0.04	1.11	40.21	59.71	0.08
R10	20.6085	106.0575	0.86	31.02	68.83	0.14	0.83	42.02	57.98	0.00	0.71	28.30	71.50	0.21	0.85	36.47	63.53	0.00
R11	20.5878	106.1298	0.81	29.70	70.20	0.09	0.79	31.81	68.19	0.00	0.79	33.97	66.00	0.02	0.61	48.60	51.31	0.09
R12	20.5197	106.1799	0.82	24.93	74.95	0.13	0.36	46.84	53.16	0.00	0.38	44.34	55.66	0.00	0.36	49.36	50.64	0.00
R13	20.4566	106.2245	0.42	46.73	53.28	0.00	0.32	58.15	41.85	0.00	0.61	44.43	55.54	0.02	0.46	53.00	47.00	0.00
R14	20.4268	106.2095	0.76	25.55	74.33	0.13	0.75	28.60	71.28	0.13	0.71	27.56	72.30	0.13	0.44	52.42	47.58	0.00
R15	20.3883	106.2861	0.60	43.37	56.54	0.09	0.67	39.50	60.40	0.10	0.49	48.55	51.45	0.00	0.78	45.79	54.14	0.07
R16	20.3264	106.2649	0.71	38.52	61.33	0.13	0.29	60.50	39.50	0.00	0.41	54.58	45.38	0.02	0.30	59.20	40.80	0.00
R17	20.3670	106.3471	0.55	25.68	74.30	0.00	0.57	54.80	45.19	0.02	0.76	40.64	59.30	0.06	1.00	47.34	52.56	0.1
R18	20.2988	106.4322	0.58	34.18	65.74	0.08	0.57	37.70	62.22	0.08	0.91	42.55	57.34	0.10	0.57	37.90	62.01	0.09
R19	20.3151	106.5294	0.67	38.45	61.46	0.08	0.50	27.40	72.60	0.00	0.77	19.06	80.82	0.13	0.35	50.83	49.17	0.00
R20	20.2540	106.5836	0.69	37.13	62.77	0.11	0.46	34.54	65.35	0.11	0.56	35.61	64.30	0.09	0.20	82.23	17.77	0.00

Annex 9: The quantifier ions used in mass spectrometry quantification, LOD values and the Vietnam national standards for priority PAHs in sediment.

Compounds	Quantifier ion (m/z)	LOD (ng g^{-1})	TEF[b]	MEF[b]	Vietnam national standards (QCVN 43:2017/BTNMT)	
					Freshwater sediment (ng g^{-1})	Brackish water and marine sediment (ng g^{-1})
Nap	128	0.02			391	391
Acy	154	0.03			128	128
Ace	152	0.07			88.9	88.9
Flu	166	0.06			144	144
Phe	178	0.10			515	544
Ant	178	0.07			245	245
Flt	202	0.07			2355	1494
Pyr	202	0.06			875	1398
BaA	228	0.05	0.1	0.082	385	693
Chr	228	0.04	0.001	0.017	862	846
BbF	252	0.12	0.1	0.25	-	-
BkF	252	0.11	0.1	0.11	-	-
BaP	252	0.17	1	1	-	-
DBahA	276	0.17	1	0.29	135	135
Ind	278	0.18	0.1	0.31	-	-
BghiP	276	0.18	0.01	0.19	-	-

[a]Carcinogenic PAHs

[b] TEF : Toxic Equivalent Factor, MEF : Mutagenic Equivalent Factor (Ambade et al., 2022; Arowojolu et al., 2021)

Annex 10: The quantifier ions used in mass spectrometry quantification, LOD values, and the Vietnamese national standards in sediment when available.

Compounds	Quantifier ion (m/z)	LOD (ng g^{-1})	Vietnam national standards (QCVN 43:2017/BTNMT; MONRE, 2017)	
			Freshwater sediment (ng g^{-1})	Brackish water and marine sediment (ng g^{-1})
Polychlorinated biphenyls (PCBs)				
PCB 28	256 → 186	1.22 (pg g^{-1})	277	189
PCB 52	290 → 220	2.40 (pg g^{-1})		
PCB 101	326 → 256	3.50 (pg g^{-1})		
PCB 118	326 → 256	5.30 (pg g^{-1})		
PCB 138	360 → 290	7.40 (pg g^{-1})		
PCB 153	360 → 290	5.60 (pg g^{-1})		
PCB 180	394 → 324	19.40 (pg g^{-1})		
Polybrominated diphenyl ethers (PBDEs)				
BDE 28	81	0.005	Not available	
BDE 47	81	0.007		
BDE 99	81	0.025		
BDE 100	81	0.004		
BDE 153	81	0.007		
BDE 154	81	0.007		
BDE 183	81	0.010		
BDE 209	79	1.01		
Chlorinated Paraffins (CPs)				
SCCPs	101 → 65	0.86	Not available	

Annex 11: The quantifier and qualifier ions used in mass spectrometry quantification, LOD values, and recovery rate of PFAS extraction methods in water and sediment samples.

Compounds	Quantifier (m/z)	Qualifier (m/z)	Recovery (%)		Recovery (%) TOPA		LOD	
			Water (n=3)	Sediment (n=3)	Water (n=3)	Sediment (n=4)	Water (ng L^{-1})	Sediment (ng g^{-1})
PFPrA	162.9824677		69.3±4.6	44.7±26.3	42.1±17	59.1±25.1	1.31	2.00
PFBA	212.979187	168.9894	109±0.9	98.9±12.1	279.7±211.8	491.5±67.9	0.09	0.10
PFPeA	262.975922	218.9862	99.8±1.6	97.2±11.6	258±184.5	442.5±73.5	0.08	0.04
PFHxA	312.972748	268.9830	98.5±1.0	101±11.7	666±507.9	1174±158.1	0.11	0.13
PFHpA	362.969147	318.9798	97.7±2.7	96.1±13.3	370.4±267.4	637.8±103	0.007	0.15
PFOA	412.966217	368.9766	99.3±3.2	94.4±15.3	1085±808	1893±277	0.03	0.87
PFNA	462.962952	418.9734	99.0±0.8	95.4±12.6	168.1±117.4	285.5±50.8	0.03	0.16
PFDA	512.959595	468.9702	98.0±2.6	96.5±10.8	145.9±89	234.9±56.9	0.13	0.13
PFUdA	562.956482	518.9670	82.2±2.8	91.2±13.7	54.2±25.8	80±28.4	1.85	0.64
PFDoA	612.953247	568.9638	101±0.8	94.6±13.8	43.7±11.4	55.1±32.3	0.05	0.36
PFTrDA	662.94989	618.9606	65.3±11.2	72.9±13.0	37.5±8.2	45.7±29.3	0.08	0.35
PFTeDA	712.946899	668.9574	99.2±2.3	107±15.2	42.1±9.4	51.5±32.7	0.15	0.44
PFODA	912.933411	868.9447	11.4±12.4	68.8±33.2	0±0	0±0	0.40	2.56
PFBS	298.942841	79.9574	93.5±1.7	86.9±22.2	351.4±237	588.4±114.4	0.46	1.87
PFPeS	348.93927	79.9574	102±2.3	96.8±11.4	46.3±40.1	86.4±6.2	0.32	0.27
PFHxS	398.93634	79.9574	99.3±1.8	99.8±12.0	252±159.4	411.4±92.6	0.01	0.18

PFHpS	448.932892	79.9574	102±2.0	99.0±12.3	55.2±36.5	91.7±18.6	0.04	0.20
PFOS	498.930115	79.9574	97.7±3.2	98.9±13.4	317.1±226.4	543.4±90.7	0.05	0.09
PFNS	548.926575	98.9558	100.5±5.0	95.0±14.7	30.3±22	52.3±8.3	0.11	0.30
PFDS	598.923279	79.9574	85.7±6.2	98.9±16.0	19±13.5	32.4±5.5	0.07	0.22
PFUnS	648.919922	98.9558	128±14.4	106.6±18.7	35.8±26	61.8±9.8	0.69	2.12
PFDoS	698.916687	98.9558	54.6±2.4	70.4±9.5	14.5±9.2	23.7±5.3	0.04	0.19
4:2-FTS	326.974091	306.9681	97.9±1.5	99.6±13.2	0±0	0±0	0.38	0.33
6:2-FTS	426.967407	80.9652	97.2±1.0	97.2±11.4	1.5±0.8	2.3±0.7	0.54	0.35
8:2-FTS	526.961182	80.9652	98.3±2.0	98.2±13.0	11.6±0	11.6±11.6	0.20	0.86
10:2-FTS	626.954651	80.9652	69.7±4.5	105.8±11.0	0±0	0±0	0.19	2.14
FBSA	297.958679	77.9655	74.1±9.5	96.4±11.9	8.7±0.3	8.4±9.1	1.78	2.40
FHxSA	397.951935	77.9655	77.3±9.9	88.6±13.9	11.6±0	11.6±11.6	0.35	2.32
FOSA	497.946106	77.9655	113.3±1.3	97.8±12.2	5.5±0.7	6.3±4.8	0.10	0.12
N-MeFOSA	511.961426	168.9894	90.9±4.3	94.4±10.5	0±0	0±0	0.05	0.15
N-EtFOSA	525.977173	218.9862	102±5.4	95.5±11.8	0±0	0±0	0.22	0.21
FOSAA	555.951416	77.9655	94.4±10.8	72.2±26.7	0±0	0±0	0.49	0.96
N-MeFOSAA	569.96759	482.9324	101.2±3.7	83.0±11.5	0±0	0±0	1.83	0.98
N-EtFOSAA	583.983093	418.9733	100±4.0	96.0±16.5	0±0	0±0	0.04	0.42
N-MeFOSE	616.008728	59.0139	98.7±1.6	94.2±10.4	0±0	0±0	1.23	1.05
N-EtFOSE	630.023926	59.0139	91.4±5.1	95.5±12.5	0±0	0±0	0.96	1.06

HPFO-DA	284.977905	168.9894	100.9±4.1	87.9±15.5	46.2±32.2	78.4±14	0.39	1.67
ADONA	376.968231	250.9760	106±1.7	75.4±11.4	0±0	0±0	1.72	0.38
PF4OPeA	228.974121	84.9906	101±1.9	99.1±12.2	105.1±77.6	182.7±27.5	0.08	0.37
PF5OHxA	278.970856	84.9906	102±2.5	101±9.5	54.4±40.5	94.9±13.9	0.18	0.45
3,6-OPFHpA	294.96582	200.9792	100±1.2	116.2±17.2	44.4±30.9	75.2±13.5	0.27	1.2
9Cl-PF3ONS	530.895081	350.9452	108±3.2	76.7±14.3	22.3±15.1	37.4±7.2	1.55	1.24
11Cl-PF3OUdS	630.888428	450.9388	83.6±8.2	74.3±7.8	11.1±8.2	19.2±2.9	1.67	0.69
PFEESA	314.937653	134.9875	92.4±1.5	98.5±12.0	46.4±38.7	85.1±7.7	0.34	0.30
3:3-FTCA	241.010437	116.9957	103.6±0.7	90.6±11.60	0±0	0±0	0.75	0.98
5:3-FTCA	341.003387	236.9956	97.5±2.3	95.8±12.4	0±0	0±0	2.50	3.25
7:3-FTCA	440.996979	316.9826	86.5±1.0	99.0±18.1	0±0	0±0	4.80	3.19
PFECHS	460.93277	380.9766	91.7±11.6	113±22.8	126.4±84.9	211.3±41.5	0.32	0.29
6:2-FTAB	569.078613	64.9730	86.5±10.1	73.5±22.6	0±0	0±0	0.22	0.98
8:2-FTUCA	456.972168	392.9766	86.4±13.1	101±11.0	0±0	0±0	0.51	1.94
6:2-diPAP	788.974365	442.9723	94.5±12.7	125±29.6	0±0	0±0	0.53	2.58
8:2-diPAP	988.961914	542.9659	130±13.9	157±33.6	0±0	0±0	0.54	2.93
10:2-diPAP	1188.94885	96.9696	10.1±13.4	144±111.2	0±0	0±0	8.8	5.15

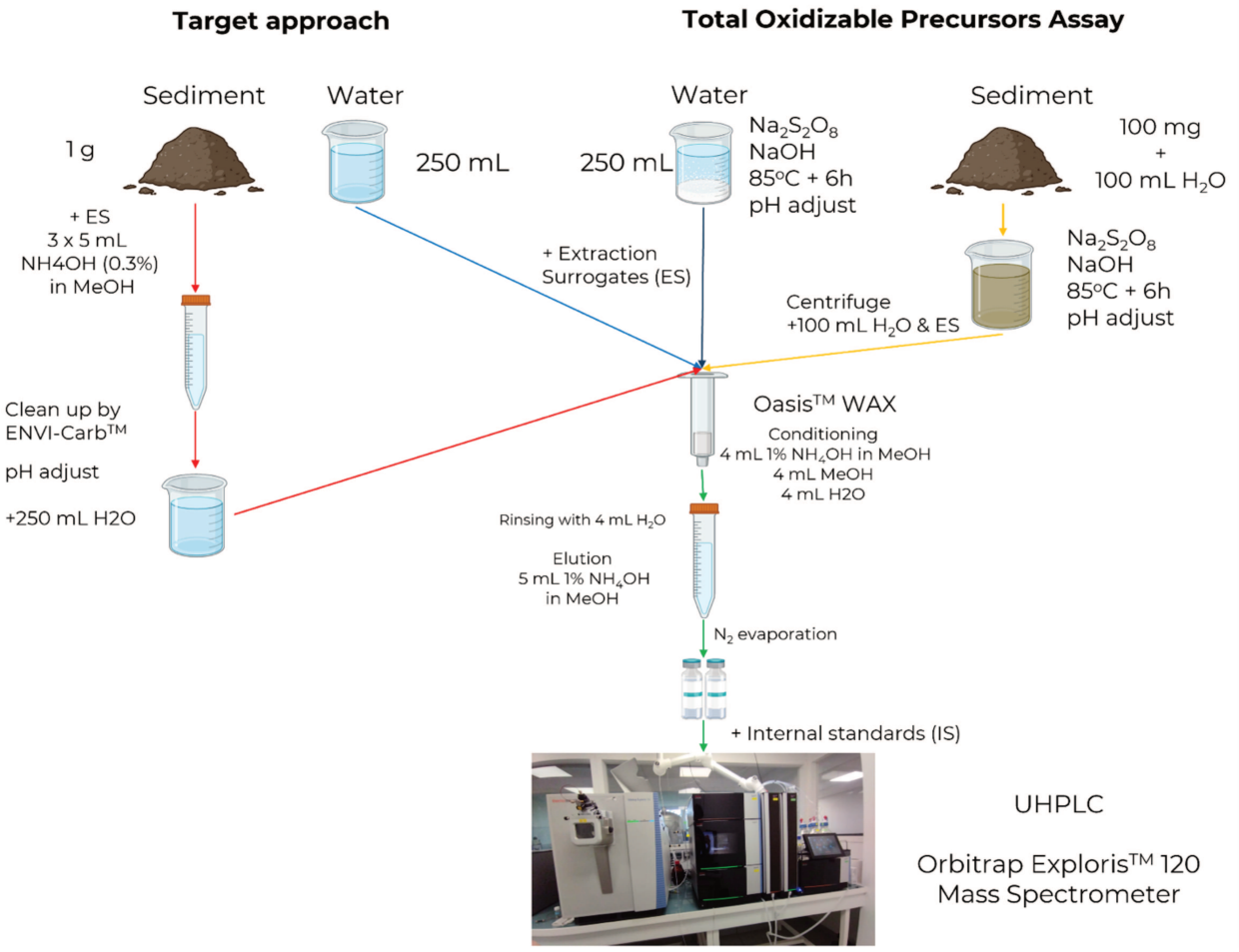
Annex 12: Extraction surrogate standards for PFAS (solution MPFAC-HIF-ES) and their recovery rates.

Labeled isotopes (surrogate)	Acronym	Concentration (ng mL^{-1})	Recovery (%)
Perfluoro-n-($^{13}C_4$)butanoic acid	MPFBA	2000	74.3±21.0
Perfluoro-n-($^{13}C_5$)pentanoic acid	M5PFPeA	1000	81.0±13.1
Perfluoro-n-(1,2,3,4,6-$^{13}C_5$)hexanoic acid	M5PFHxA	500	76.3±8.4
Perfluoro-n-(1,2,3,4-$^{13}C_4$)heptanoic acid	M4PFHpA	500	79.9±9.3
Perfluoro-n-($^{13}C_8$)octanoic acid	M8PFOA	500	85.4±9.6
Perfluoro-n-($^{13}C_9$)nonanoic acid	M9PFNA	250	86.9±9.0
Perfluoro-n-(1,2,3,4,5,6-$^{13}C_6$)decanoic acid	M6PFDA	250	84.1±8.9
Perfluoro-n-(1,2,3,4,5,6,7-$^{13}C_7$)undecanoic acid	M7PFUdA	250	92.3±16.4
Perfluoro-n-(1,2-$^{13}C_2$)dodecanoic acid	M2PFDoA	250	79.8±15.9
Perfluoro-n-(1,2-$^{13}C_2$)tetradecanoic acid	M2PFTeDA	250	52.2±17.8
Perfluoro-1-($^{13}C_8$)octanesulfonamide	M8FOSA	500	77.9±9.1
N-Methyl-d_3-perfluoro-1-octanesulfonamide	d-N-MeFOSA	500	56.4±12.6
N-Ethyl-d_5-perfluoro-1-octanesulfonamide	d-N-EtFOSA	500	49.9±13.1
2-(N-Methyl-d_3-perfluorooctane sulfonamido)acetic acid	d3-N-MeFOSAA	1000	90.7±26.7
N-Ethyl-d_5-perfluorooctane sulfonamidoacetic acid	d5-N-EtFOSAA	1000	75.6±11.4
N-Methyl-d_3-perfluorooctane sulfonamidoethan-d_4-ol	d7-N-MeFOSE	5000	46.3±12.2
N-Ethyl-d_5-perfluorooctane sulfonamidoethan-d_4-ol	d9-N-EtFOSE	5000	42.2±13.9
2,3,3,3-tetrafluoro-2-(heptafluoropropoxy)($^{13}C_3$)propionic acid	M3HPFO-DA	2000	112.7±27.4
Perfluoro-1-(2,3,4-$^{13}C_3$)butanesulfonic acid	M3PFBS	466	83.6±9.2
Perfluoro-1-(2,3,4-$^{13}C_3$)hexanesulfonic acid	M3PFHxS	474	85.6±9.5
Perfluoro-1-($^{13}C_8$)octanesulfonic acid	M8PFOS	479	81.6±8.1
1H,1H,2H,2H-Perfluoro(1,2-$^{13}C_2$)hexanesulfonic acid	M2-4:2-FTS	938	90.2±8.9
1H,1H,2H,2H-perfluoro(1,2-$^{13}C_2$)octanesulfonic acid	M2-6:2-FTS	951	87.0±8.5
1H,1H,2H,2H-perfluoro(1,2-$^{13}C_2$)decanesulfonic acid	M2-8:2 FTS	960	83.4±11.8

Annex 13: Injection internal standards (solution MPFAC-HIF-IS).

Labeled isotopes (internal standards)	Acronym	Concentration (ng mL^{-1})
Perfluoro-n-(2,3,4-$^{13}C_3$)butanoic acid	M3PFBA	1000
Perfluoro-n-(1,2-$^{13}C_2$)hexanoic acid	MPFHxA	500
Perfluoro-n-(1,2,3,4-$^{13}C_4$)octanoic acid	MPFOA	500
Perfluoro-n-(1,2,3,4,5-$^{13}C_5$)nonanoic acid	MPFNA	250
Perfluoro-n-(1,2-$^{13}C_2$)decanoic acid	MPFDA	250
Perfluoro-1-hexane-($^{18}O_2$)sulfonic acid	MPFHxS	474
Perfluoro-1-(1,2,3,4-$^{13}C_4$)octanesulfonic acid	MPFOS	479

Annex 14: General schema of PFAS extraction methods for water and sediment samples



Annex 15: The chromatography and mass spectrometry conditions used for the analysis of PCBs, PBDEs, and SCCPs adapted from Godéré et al. (2022) and Rigal et al. (2024).

	PCBs	CPs	PBDEs
Column	HP-5MS (Agilent) 30m x 0.25mm x 0.25µM		RTX-1614 column (Restek) 15m x 0.25 mm x 0.1µm
Injection volume	1 µL		
Injector temperature (°C)	300	280	260
Transfer line temperature (°C)	300	300	280
Injection mode	Pulsed spitless		
Source temperature	280	250	250
Ionization Mode	Electronic Impact (70 eV)	Electronic Impact (70 eV)	Electronic Capture Negative Ionization (N_2)
Oven program	120°C (hold 2 min) to 260°C (at 10°C.min^{-1}), then to 300°C (at 20°C.min^{-1} and hold for 6 min)	70 °C (hold 1 min) to 300°C (at 15°C.min^{-1} and hold for 8 min)	70 °C (hold 4 min) to 220°C at 10°C.min^{-1} and hold 6 min), then to 230°C (at 5°C.min^{-1}) and to 315°C (at 10°C.min^{-1} and hold 5 min)
Acquisition mode	Multiple Reaction Monitoring (MRM)		Single Ion Monitoring (SIM)

Annex 16: MS conditions for PFAS analysis.

Parameters	Settings
Run time	19 min
Ion source	H-ESI
Spray voltage	Negative mode, 2.5 kV
Sheath gas (Arb)	50
Auxiliary gas (Arb)	10
Sweep gas (Arb)	1
Ion Transfer Tube Temperature (^{o}C)	225
Vaporize Temperature (^{o}C)	350
Acquisition mode	Full Scan with data-independent acquisition (DIA)
Chromatography peak width (s)	6
Full Scan Orbitrap Resolution	60000
Full Scan Range (m/z)	100 - 1000
RF Lens (%)	70
DIA precursor mass range (m/z)	50 - 1200
HCD collision energies (%)	Stepped 10,50
DIA Orbitrap Resolution	15000
DIA m/z windows	12 x 105 m/z windows
Mass extraction	5 ppm

Annex

Annex 17: Concentration (ng g^{-1}) of the 16 PAHs in the Red River sediments from the three sampling campaigns in 2023

	R00			R01			R02			R03			R04		
Month	03	06	09	03	06	09	03	06	09	03	06	09	03	06	09
Nap	*<LOD*	*1.53*	*<LOD*	*0.74*	*1.14*	*1.12*	*0.32*	*5.06*	*0.46*	*<LOD*	*1.91*	*0.37*	*<LOD*	*1.80*	*0.02*
Acy	*0.13*	*1.13*	*0.55*	*0.66*	*0.71*	*0.76*	*0.76*	*1.15*	*0.71*	*0.19*	*0.53*	*0.42*	*0.09*	*0.30*	*0.14*
Ace	*<LOD*	*0.39*	*0.14*	*<LOD*	*0.10*	*0.13*	*0.11*	*0.23*	*0.18*	*0.18*	*0.23*	*0.09*	*<LOD*	*0.31*	*0.15*
Flu	*0.43*	*1.68*	*0.75*	*0.92*	*0.78*	*1.00*	*1.03*	*1.56*	*0.83*	*1.36*	*1.14*	*1.04*	*0.83*	*2.40*	*0.34*
Phe	*2.85*	*10.85*	*9.29*	*6.43*	*8.21*	*9.56*	*7.79*	*12.34*	*8.98*	*10.19*	*7.38*	*9.02*	*4.86*	*14.82*	*4.06*
Ant	*0.99*	*1.21*	*1.21*	*1.45*	*0.97*	*0.62*	*1.68*	*1.43*	*0.69*	*1.56*	*0.89*	*0.68*	*1.29*	*1.29*	*0.08*
Flt	*1.82*	*9.26*	*10.45*	*6.59*	*8.28*	*10.77*	*10.26*	*10.48*	*9.60*	*10.74*	*6.54*	*9.23*	*4.04*	*16.67*	*4.48*
Pyr	*0.21*	*8.40*	*9.37*	*4.50*	*6.34*	*9.85*	*6.97*	*8.99*	*10.88*	*6.88*	*5.26*	*8.36*	*2.08*	*11.74*	*3.28*
BaA	*2.40*	*3.19*	*2.67*	*3.73*	*3.43*	*3.42*	*4.47*	*3.56*	*3.92*	*4.65*	*2.42*	*3.00*	*3.11*	*5.59*	*2.41*
Chr	*0.35*	*5.58*	*5.41*	*1.70*	*4.86*	*2.47*	*3.04*	*5.70*	*3.10*	*3.45*	*4.09*	*5.17*	*0.83*	*9.66*	*2.06*
BbF	*3.61*	*7.86*	*7.68*	*5.98*	*7.23*	*6.83*	*8.30*	*9.05*	*8.74*	*8.79*	*6.06*	*7.79*	*4.82*	*13.93*	*7.88*
BkF	*<LOD*	*2.11*	*2.92*	*<LOD*	*2.07*	*1.06*	*0.28*	*2.74*	*1.09*	*0.27*	*1.41*	*2.18*	*<LOD*	*3.78*	*0.37*
BaP	*0.38*	*2.76*	*2.34*	*1.03*	*2.75*	*<LOD*	*2.31*	*3.46*	*2.68*	*1.65*	*1.85*	*2.74*	*0.52*	*3.74*	*0.62*
DBahA	*0.82*	*1.65*	*1.18*	*1.17*	*1.46*	*0.55*	*1.18*	*1.54*	*<LOD*	*1.25*	*1.41*	*1.24*	*1.13*	*2.29*	*0.71*
Ind	*1.19*	*3.51*	*2.70*	*2.05*	*2.75*	*2.10*	*2.89*	*3.52*	*2.74*	*2.27*	*2.13*	*2.52*	*1.66*	*5.02*	*1.45*
BghiP	*1.01*	*4.60*	*3.72*	*2.26*	*3.64*	*2.81*	*3.51*	*5.59*	*4.52*	*2.94*	*2.60*	*3.26*	*1.52*	*1.36*	*1.64*

Annex

	R5			R6			R7			R8			R9		
Month	03	06	09	03	06	09	03	06	09	03	06	09	03	06	09
Nap	*<LOD*	*0.52*	*0.08*	*<LOD*	*3.27*	*0.20*	*0.13*	*4.55*	*0.03*	*0.22*	*3.56*	*0.29*	*0.21*	*<LOD*	*0.95*
Acy	*0.19*	*0.61*	*0.28*	*0.07*	*0.59*	*0.30*	*0.32*	*0.98*	*0.23*	*0.38*	*1.18*	*0.56*	*0.41*	*0.15*	*0.71*
Ace	*0.08*	*0.26*	*0.09*	*<LOD*	*0.18*	*0.17*	*0.25*	*0.33*	*0.12*	*0.08*	*0.22*	*0.15*	*0.11*	*<LOD*	*0.31*
Flu	*1.20*	*1.35*	*1.25*	*0.73*	*1.18*	*0.76*	*0.85*	*1.35*	*0.47*	*0.97*	*1.55*	*0.93*	*1.16*	*0.77*	*1.41*
Phe	*7.24*	*10.49*	*9.72*	*4.19*	*7.66*	*6.36*	*5.62*	*13.19*	*4.37*	*7.31*	*12.53*	*16.99*	*6.73*	*4.94*	*12.71*
Ant	*1.78*	*1.35*	*0.90*	*1.34*	*0.87*	*0.38*	*1.44*	*1.05*	*0.30*	*1.61*	*1.47*	*0.63*	*1.46*	*0.62*	*1.96*
Flt	*10.69*	*9.21*	*10.68*	*6.22*	*6.48*	*6.41*	*5.89*	*10.28*	*3.61*	*9.54*	*10.99*	*21.97*	*8.45*	*4.84*	*13.56*
Pyr	*7.49*	*7.42*	*9.12*	*3.25*	*5.30*	*5.06*	*4.07*	*9.64*	*3.07*	*5.88*	*9.34*	*33.69*	*4.51*	*3.60*	*13.19*
BaA	*5.03*	*3.27*	*2.72*	*3.42*	*2.59*	*3.13*	*3.56*	*3.37*	*2.52*	*4.17*	*3.77*	*5.56*	*4.06*	*2.07*	*5.26*
Chr	*3.68*	*5.43*	*5.93*	*0.72*	*3.81*	*2.01*	*1.19*	*6.31*	*1.61*	*1.55*	*6.78*	*2.65*	*2.72*	*2.98*	*6.79*
BbF	*9.89*	*7.71*	*7.40*	*5.50*	*5.45*	*6.53*	*5.72*	*8.03*	*5.03*	*8.65*	*9.18*	*15.43*	*8.47*	*4.79*	*14.10*
BkF	*<LOD*	*1.68*	*1.84*	*<LOD*	*1.68*	*1.01*	*<LOD*	*1.99*	*0.80*	*0.21*	*2.22*	*1.05*	*0.23*	*1.15*	*2.74*
BaP	*2.31*	*2.77*	*1.74*	*0.76*	*2.10*	*1.17*	*1.14*	*2.74*	*0.83*	*1.89*	*3.19*	*2.00*	*1.57*	*1.51*	*3.20*
DBahA	*1.35*	*1.42*	*1.23*	*1.17*	*1.41*	*0.59*	*1.13*	*1.70*	*0.41*	*1.21*	*1.57*	*0.55*	*1.23*	*1.46*	*1.64*
Ind	*2.93*	*2.53*	*2.09*	*1.63*	*2.36*	*1.40*	*1.81*	*3.32*	*1.12*	*2.65*	*3.60*	*2.17*	*2.58*	*1.65*	*4.39*
BghiP	*4.01*	*3.44*	*2.87*	*1.68*	*2.65*	*2.10*	*1.81*	*4.16*	*1.52*	*3.25*	*5.24*	*3.40*	*2.89*	*1.69*	*6.03*

Annex

	R10			R11			R12			R13			R14		
Month	03	06	09	03	06	09	03	06	09	03	06	09	03	06	09
Nap	*<LOD*	*4.74*	*1.03*	*<LOD*	*3.52*	*0.04*	*<LOD*	*<LOD*	*<LOD*	*<LOD*	*2.55*	*<LOD*	*<LOD*	*4.77*	*0.10*
Acy	*0.08*	*0.50*	*0.39*	*0.09*	*0.66*	*0.12*	*0.04*	*0.07*	*0.11*	*0.09*	*0.41*	*0.08*	*0.18*	*0.88*	*0.14*
Ace	*0.09*	*0.20*	*0.17*	*0.08*	*0.29*	*0.09*	*0.09*	*0.11*	*0.12*	*0.08*	*0.16*	*0.07*	*0.08*	*0.38*	*0.12*
Flu	*0.94*	*2.18*	*1.03*	*0.72*	*1.59*	*0.74*	*0.48*	*0.45*	*0.14*	*0.61*	*1.30*	*0.29*	*0.80*	*2.14*	*0.66*
Phe	*7.40*	*12.97*	*5.94*	*4.21*	*12.25*	*4.59*	*2.73*	*3.40*	*3.78*	*3.22*	*7.02*	*4.42*	*6.75*	*16.17*	*4.71*
Ant	*1.60*	*0.88*	*0.60*	*1.62*	*1.15*	*0.38*	*1.33*	*0.45*	*0.16*	*1.32*	*0.82*	*0.18*	*1.54*	*1.70*	*0.25*
Flt	*5.62*	*8.14*	*5.28*	*4.80*	*9.31*	*3.94*	*2.73*	*3.23*	*7.11*	*2.81*	*6.01*	*3.63*	*6.35*	*15.76*	*4.85*
Pyr	*3.22*	*6.83*	*4.07*	*3.57*	*8.15*	*2.66*	*2.04*	*2.48*	*7.99*	*2.14*	*5.14*	*3.02*	*5.27*	*12.66*	*4.43*
BaA	*3.33*	*3.00*	*3.31*	*4.49*	*3.26*	*2.20*	*3.71*	*1.77*	*3.58*	*3.49*	*2.50*	*0.80*	*4.35*	*5.36*	*1.60*
Chr	*1.35*	*4.74*	*1.96*	*1.39*	*5.20*	*1.60*	*0.65*	*2.42*	*2.16*	*0.42*	*3.67*	*0.82*	*2.43*	*8.19*	*1.27*
BbF	*4.82*	*7.19*	*7.47*	*7.34*	*8.03*	*6.36*	*6.07*	*4.25*	*9.56*	*5.61*	*6.74*	*2.70*	*7.75*	*11.29*	*4.24*
BkF	*<LOD*	*1.66*	*0.63*	*<LOD*	*1.92*	*0.57*	*<LOD*	*0.86*	*0.98*	*<LOD*	*1.48*	*0.35*	*<LOD*	*3.39*	*0.54*
BaP	*0.52*	*2.08*	*0.80*	*1.32*	*2.46*	*0.43*	*1.23*	*1.29*	*1.33*	*0.68*	*2.15*	*0.32*	*1.23*	*3.11*	*0.53*
DBahA	*1.12*	*1.63*	*0.28*	*1.45*	*1.69*	*<LOD*	*1.28*	*1.21*	*0.52*	*1.24*	*1.42*	*<LOD*	*1.24*	*1.63*	*0.28*
Ind	*1.54*	*2.07*	*0.91*	*2.44*	*2.48*	*0.52*	*2.01*	*1.27*	*1.15*	*1.83*	*2.23*	*0.39*	*2.35*	*2.88*	*0.58*
BghiP	*1.13*	*2.67*	*1.24*	*2.47*	*3.55*	*0.75*	*1.99*	*1.22*	*0.98*	*1.91*	*2.88*	*0.40*	*2.45*	*4.19*	*0.63*

Annex

	R15			R16			R17			R18			R19		
Month	03	06	09	03	06	09	03	06	09	03	06	09	03	06	09
Nap	*<LOD*	*1.92*	*1.06*	*<LOD*	*1.50*	*<LOD*	*<LOD*	*0.25*	*<LOD*	*0.33*	*0.94*	*0.22*	*<LOD*	*3.15*	*0.06*
Acy	*0.10*	*0.22*	*0.35*	*0.07*	*0.21*	*0.11*	*0.06*	*0.14*	*0.17*	*0.16*	*0.34*	*0.12*	*0.07*	*0.41*	*0.14*
Ace	*0.11*	*0.13*	*0.13*	*0.10*	*0.13*	*0.10*	*<LOD*	*0.10*	*0.10*	*0.11*	*0.22*	*0.11*	*<LOD*	*0.18*	*0.16*
Flu	*0.68*	*0.85*	*1.66*	*0.59*	*0.80*	*0.31*	*0.66*	*0.94*	*0.79*	*0.96*	*2.17*	*0.75*	*0.81*	*1.76*	*0.41*
Phe	*3.94*	*4.76*	*9.22*	*3.25*	*3.95*	*3.60*	*2.91*	*7.39*	*6.79*	*4.95*	*9.00*	*4.15*	*4.06*	*9.23*	*3.59*
Ant	*1.25*	*0.56*	*0.77*	*1.47*	*0.52*	*<LOD*	*1.76*	*0.84*	*0.44*	*1.42*	*1.14*	*0.28*	*1.46*	*1.05*	*0.07*
Flt	*4.59*	*4.31*	*10.22*	*3.36*	*3.74*	*5.12*	*2.15*	*6.54*	*6.33*	*5.48*	*10.34*	*3.61*	*0.70*	*7.38*	*6.13*
Pyr	*3.05*	*3.84*	*8.68*	*1.23*	*2.97*	*5.97*	*1.04*	*5.54*	*4.75*	*3.83*	*8.34*	*3.21*	*2.78*	*6.14*	*4.93*
BaA	*3.70*	*1.94*	*2.05*	*3.37*	*1.89*	*1.10*	*3.78*	*2.73*	*4.23*	*3.67*	*3.20*	*1.22*	*3.46*	*1.95*	*1.34*
Chr	*1.22*	*2.89*	*3.85*	*<LOD*	*2.59*	*0.90*	*<LOD*	*4.13*	*2.84*	*1.25*	*5.05*	*1.05*	*0.38*	*3.57*	*1.03*
BbF	*6.14*	*4.39*	*4.99*	*4.25*	*4.09*	*2.64*	*4.88*	*7.72*	*12.00*	*6.18*	*7.53*	*4.45*	*4.93*	*4.86*	*6.80*
BkF	*<LOD*	*1.15*	*1.37*	*<LOD*	*1.01*	*0.59*	*<LOD*	*1.44*	*0.93*	*<LOD*	*2.08*	*0.36*	*<LOD*	*1.20*	*0.62*
BaP	*0.87*	*1.59*	*1.32*	*0.76*	*1.46*	*0.43*	*0.80*	*2.16*	*0.91*	*0.98*	*2.46*	*0.52*	*0.91*	*1.68*	*0.77*
DBahA	*1.14*	*1.34*	*0.94*	*1.25*	*1.35*	*<LOD*	*1.51*	*1.50*	*0.46*	*1.19*	*1.40*	*<LOD*	*1.21*	*1.29*	*<LOD*
Ind	*1.75*	*1.51*	*1.41*	*1.76*	*1.46*	*0.62*	*2.02*	*2.03*	*1.37*	*1.94*	*2.48*	*0.53*	*1.66*	*1.58*	*1.08*
BghiP	*1.76*	*1.75*	*1.85*	*1.45*	*1.57*	*0.64*	*1.64*	*2.78*	*1.92*	*1.81*	*3.13*	*0.66*	*1.54*	*1.87*	*1.12*

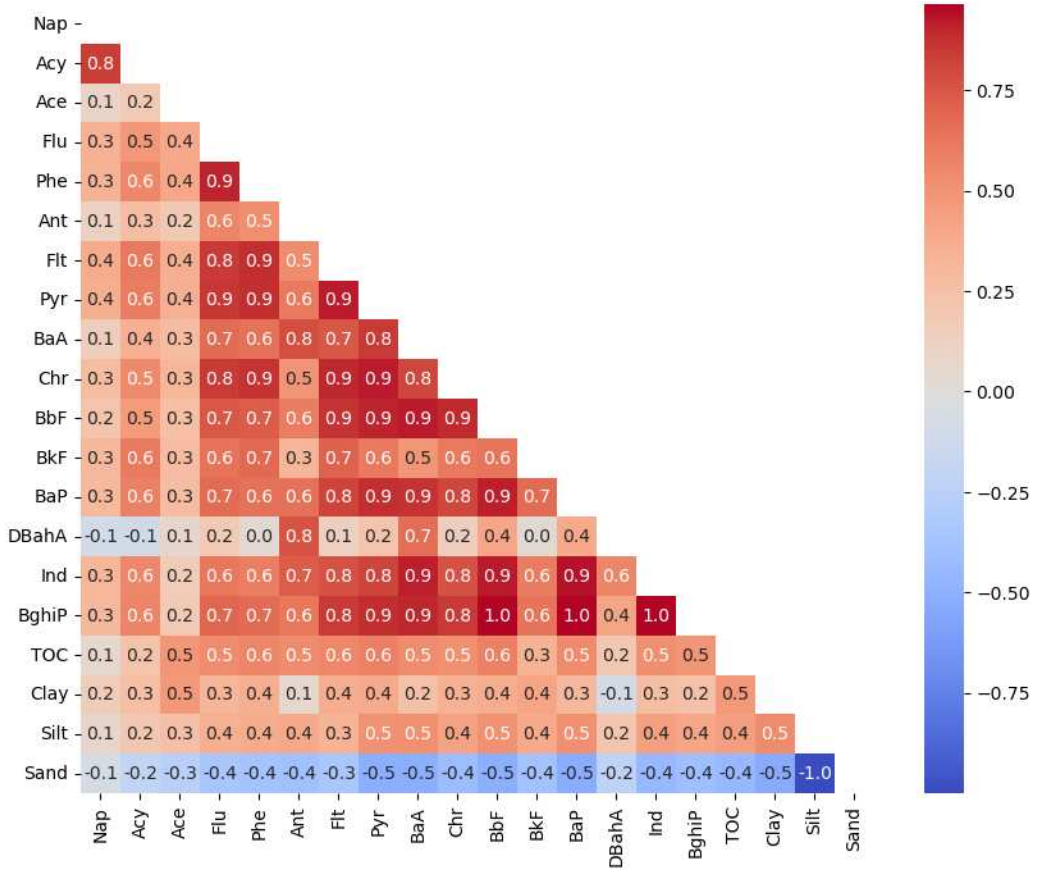
Annex

	R20		
Month	03	06	09
Nap	*<LOD*	*2.33*	*<LOD*
Acy	*0.09*	*0.31*	*<LOD*
Ace	*0.10*	*0.18*	*<LOD*
Flu	*0.70*	*1.78*	*0.26*
Phe	*3.42*	*8.06*	*2.44*
Ant	*1.30*	*1.23*	*0.24*
Flt	*2.98*	*6.59*	*5.62*
Pyr	*1.88*	*4.66*	*7.97*
BaA	*2.96*	*2.18*	*0.77*
Chr	*0.26*	*2.90*	*1.08*
BbF	*4.39*	*4.61*	*1.78*
BkF	*<LOD*	*0.84*	*0.41*
BaP	*0.74*	*1.48*	*0.78*
DBahA	*1.11*	*1.23*	*0.85*
Ind	*1.55*	*1.43*	*0.68*
BghiP	*1.27*	*1.72*	*0.49*

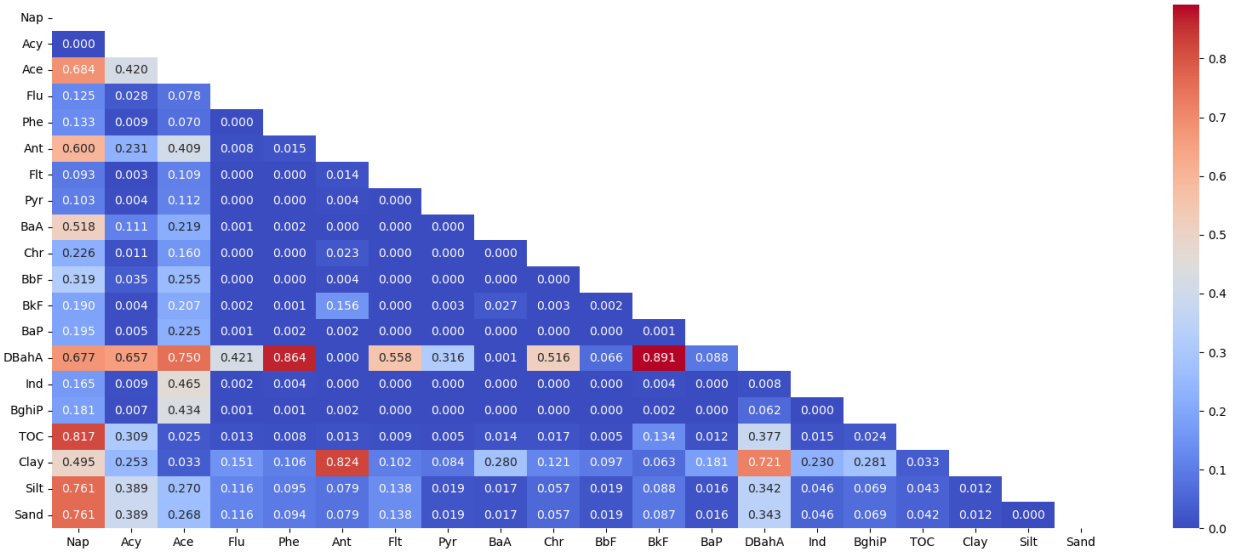
Annex

Annex 18: The a) correlation and b) p-value matrices for the concentration of PAHs and the sediment physicochemical parameters of samples in March 2023

a

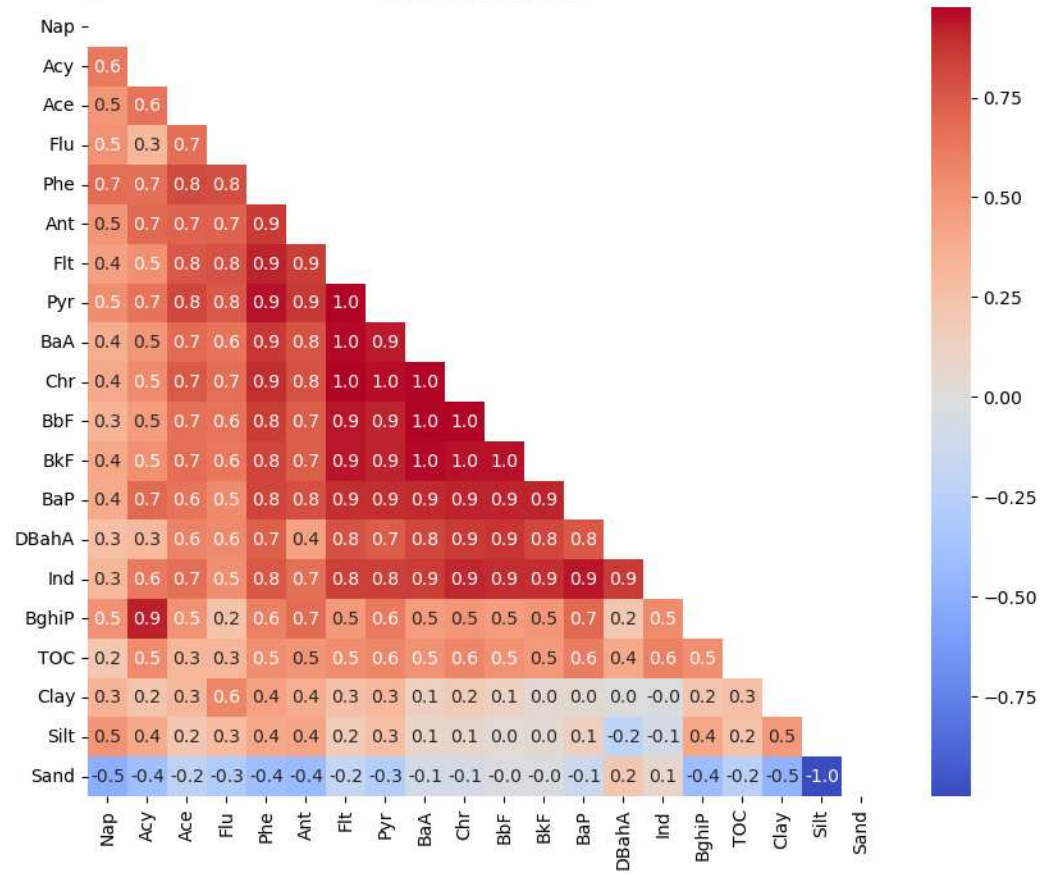


b



Annex 19: The a) correlation and b) p-value matrices for the concentration of PAHs and the sediment physicochemical parameters of samples in June 2023

a



b

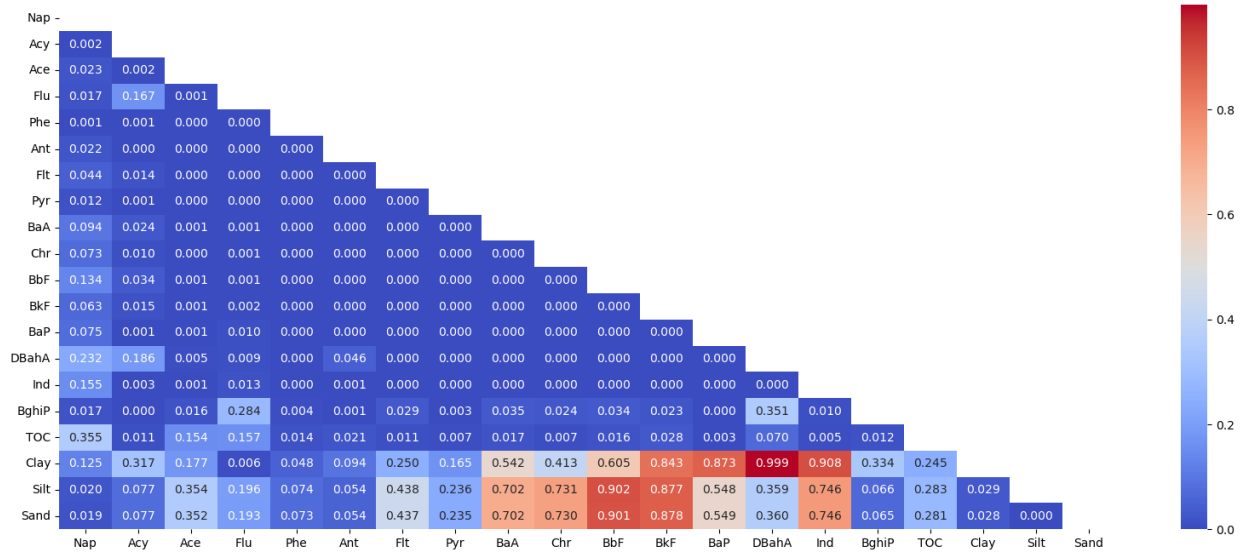
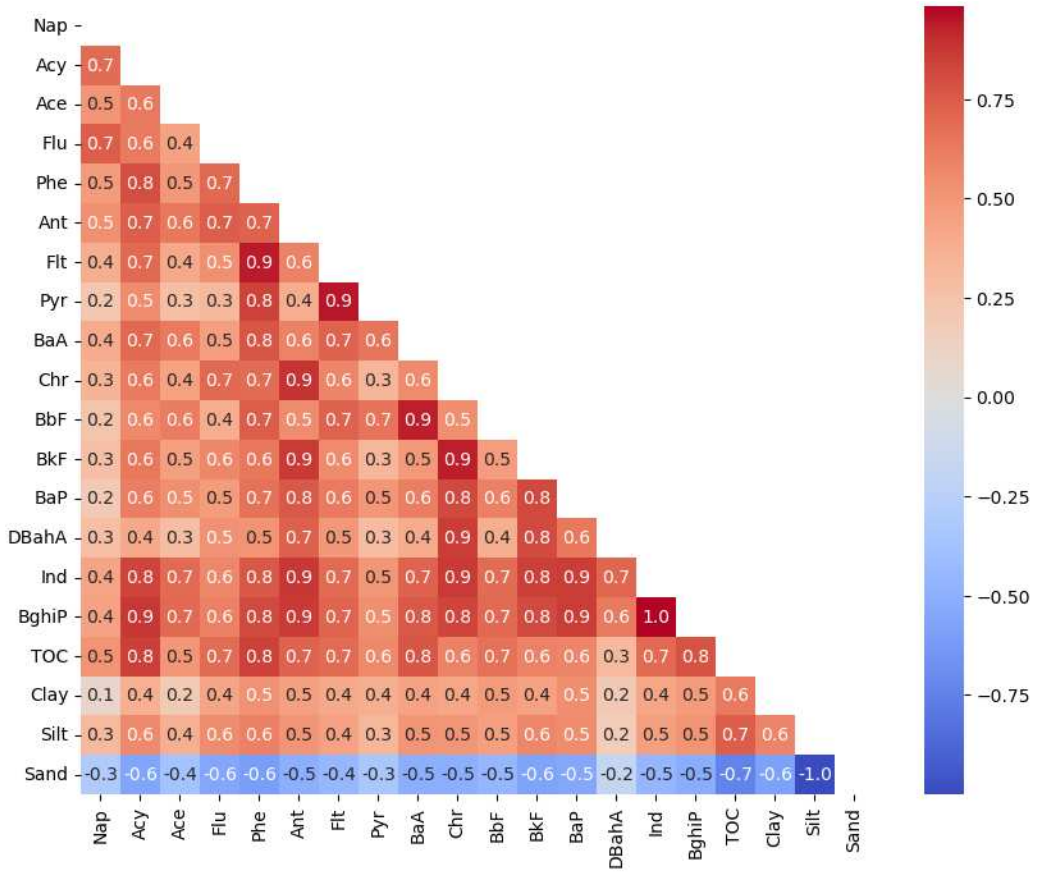


Figure S1: The a) correlation and b) p-value matrices for the concentration of PAHs and the sediment physicochemical parameters of samples in June 2023

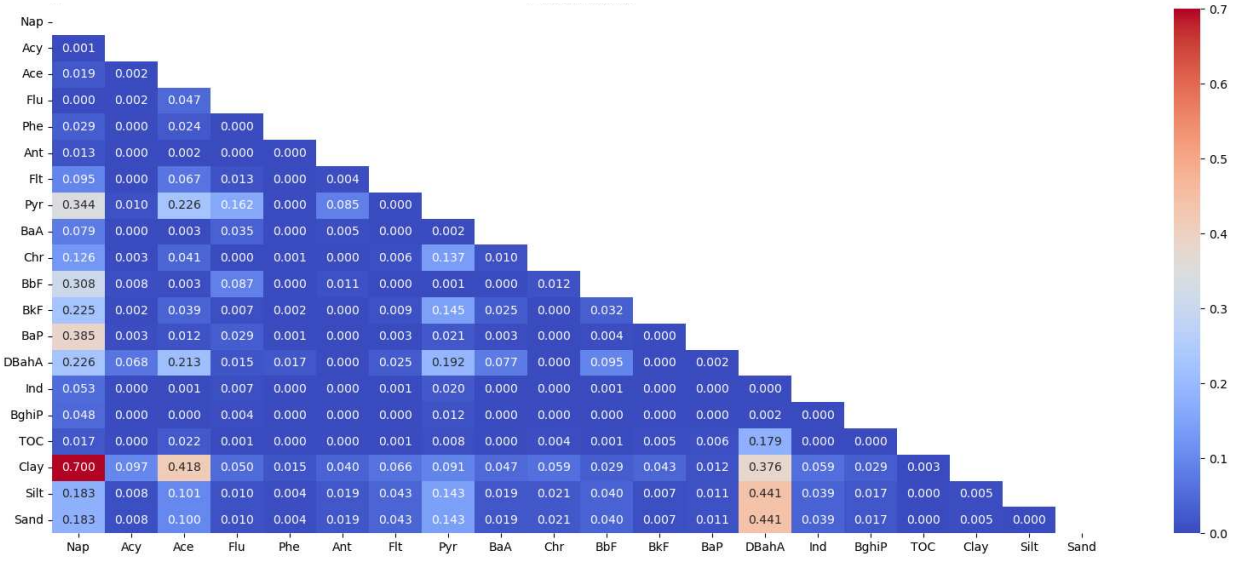
Annex

Annex 20: The a) correlation and b) p-value matrices for the concentration of PAHs and the sediment physicochemical parameters of samples in September 2023

a

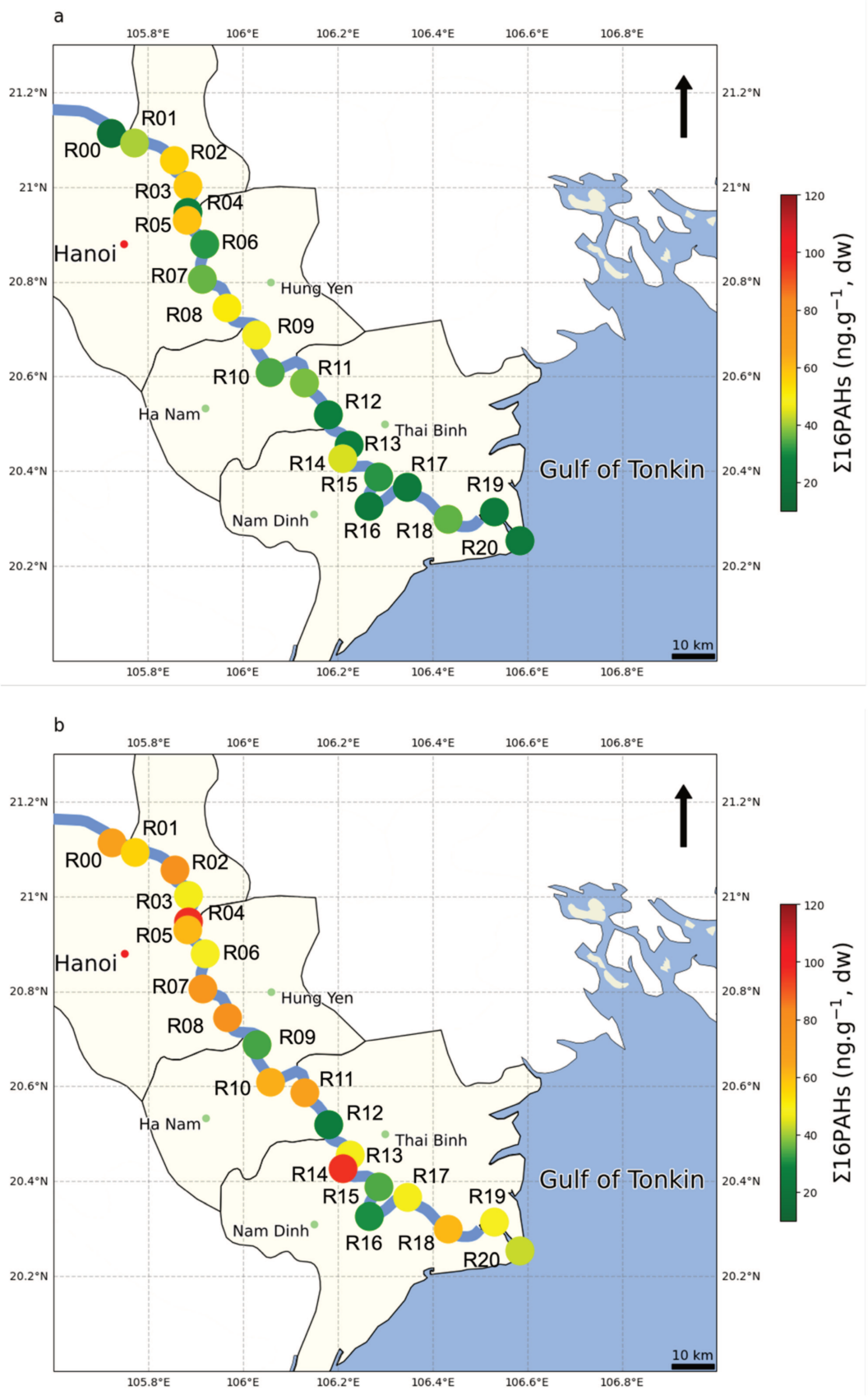


b

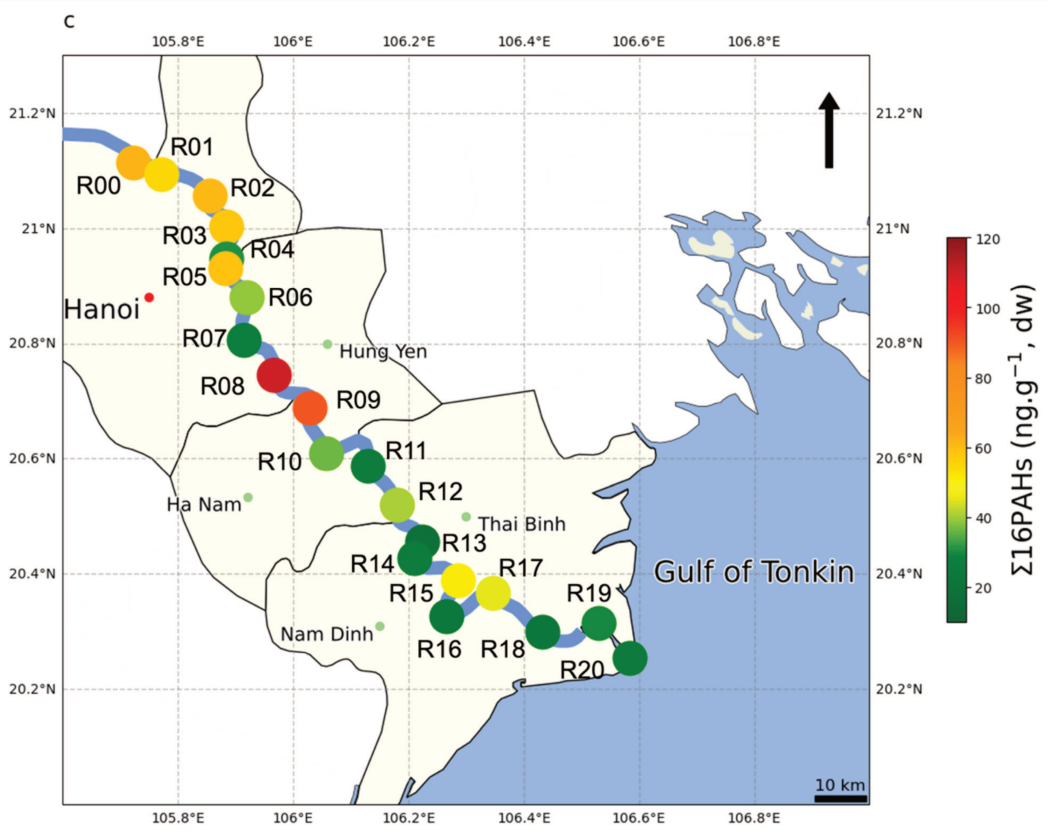


Annex

Annex 21: Spatial distribution of ∑PAHs along the Red River: a) March 2023; b) June 2023; c) September 2023.



Annex


c
105.8°E
106°E
106.2°E
106.4°E
106.6°E
106.8°E
21.2°N
21°N
20.8°N
20.6°N
20.4°N
20.2°N
R00
R01
R02
R03
R04
R05
R06
R07
R08
R09
R10
R11
R12
R13
R14
R15
R16
R17
R18
R19
R20
Hanoi
Hung Yen
Ha Nam
Thai Binh
Nam Dinh
Gulf of Tonkin
10 km
120
100
80
60
40
20
Σ16PAHs (ng.g⁻¹, dw)

Annex 22: Student's t-test result for the comparison of the mean ∑16PAHs between March and June 2023 samples.

	March 2023	*June 2023*
Mean	34.96095	55.98678
Variance	149.9252	389.3151
Observations	21	21
Pooled Variance	269.6201	
Hypothesized Mean Difference	0	
df	40	
t Stat	-4.14927	
P(T<=t) one-tail	**8.46E-05**	
t Critical one-tail	1.683851	
P(T<=t) two-tail	0.000169	
t Critical two-tail		

Annex 23: Student's t-test result for the comparison of the mean ∑16PAHs between March and September 2023 samples.

	March 2023	*September 2023*
Mean	34.96095	43.28424
Variance	149.9252	542.6107
Observations	21	21
Pooled Variance	346.2679	
Hypothesized Mean Difference	0	
df	40	
t Stat	-1.44938	
P(T<=t) one-tail	**0.077513**	
t Critical one-tail	1.683851	
P(T<=t) two-tail	0.155026	
t Critical two-tail	2.021075	

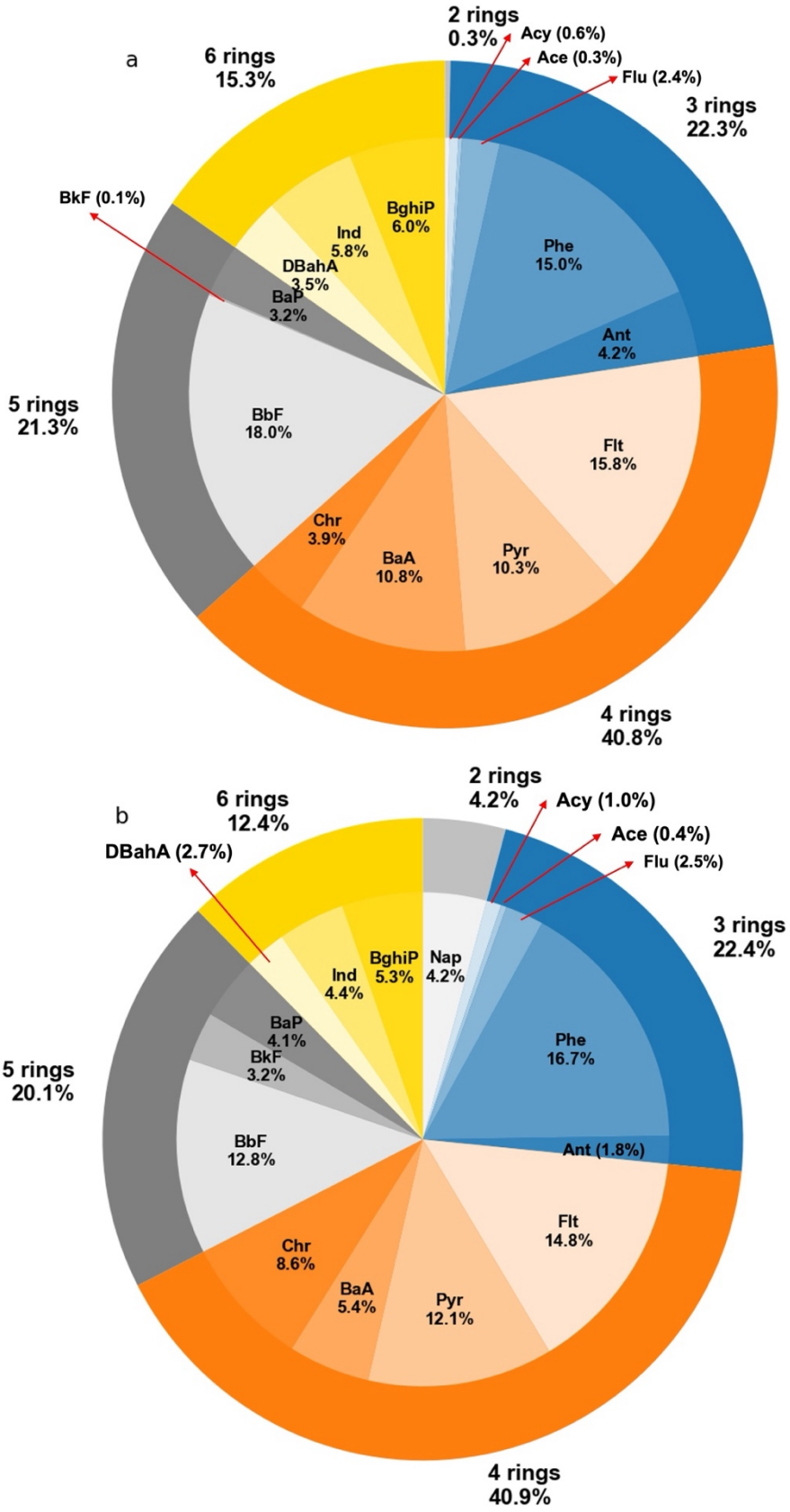
Annex 24: Student's t-test result for the comparison of the mean ∑16PAHs between June and September 2023 samples.

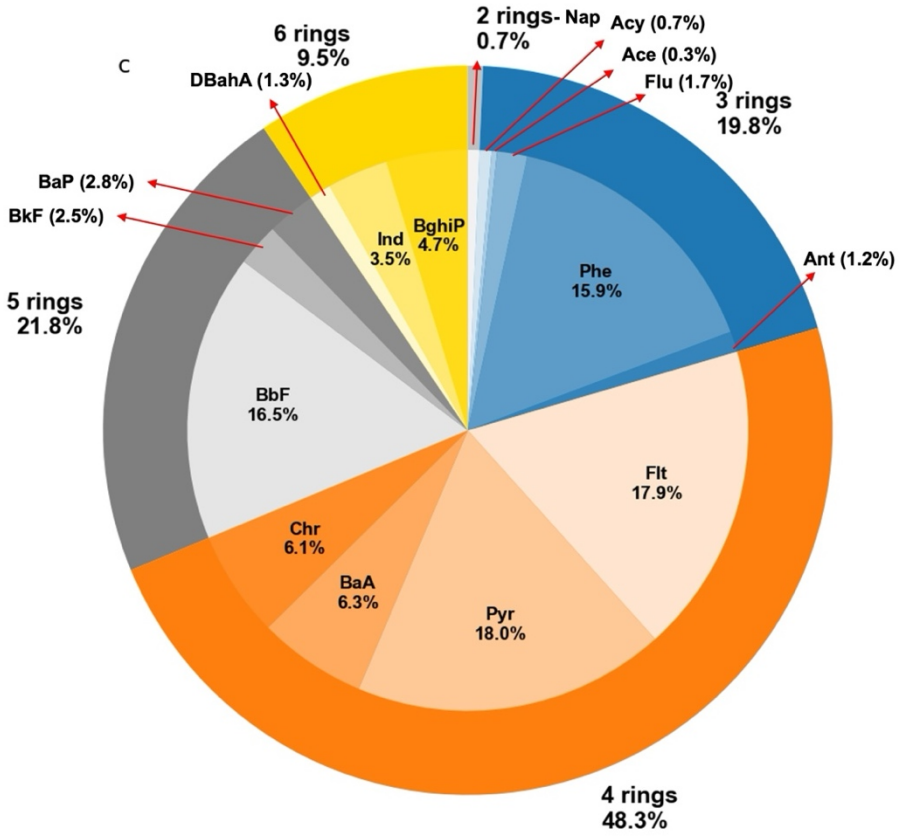
	June 2023	*September 2023*
Mean	55.98678	43.28424
Variance	389.3151	542.6107
Observations	21	21
Pooled Variance	465.9629	
Hypothesized Mean Difference	0	
df	40	
t Stat	1.906819	

Annex

P(T<=t) one-tail	**0.031872**
t Critical one-tail	1.683851
P(T<=t) two-tail	0.063745
t Critical two-tail	2.021075

Annex 25: The contribution of individual PAHs according to their ring number in the 21 sediment samples in a) March, b) June, and c) September 2023.





Annex 26: Main PAH molecular diagnostic ratios used in the literature.

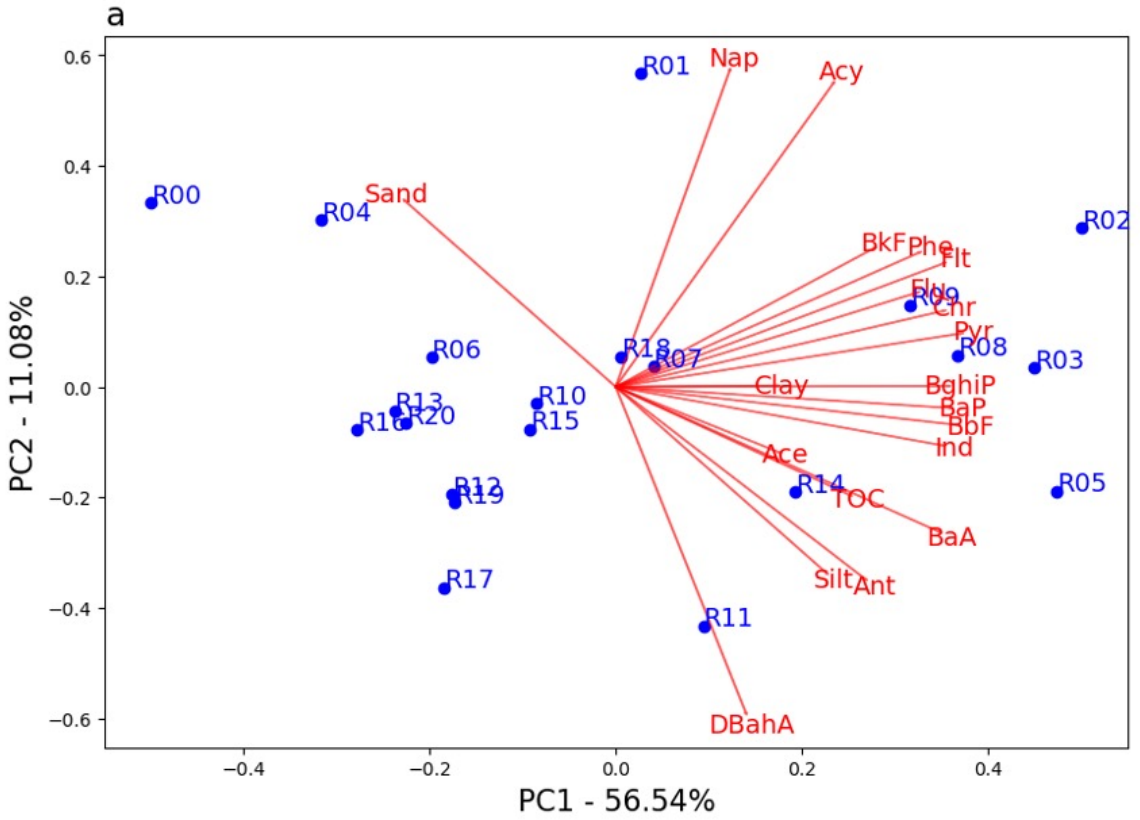
Ratios	Signification	Reference
LMW/HMW	>1: Petrogenic	(Giuliani et al., 2008; Hung et al., 2015; Kanzari et al., 2014; Pérez-Fernández et al., 2018)
BaA/(BaA+Chr)	<0.2: Petrogenic >0.35: Pyrogenic	
Ind/(Ind+BghiP)	<0.2: Petrogenic 0.2 – 0.5: Fossil fuel combustion >0.5: Biomass combustion	
Flt/(Flt+Pyr)	<0.4: petrogenic 0.4-0.5: fossil fuel combustion >0.5: pyrogenic	

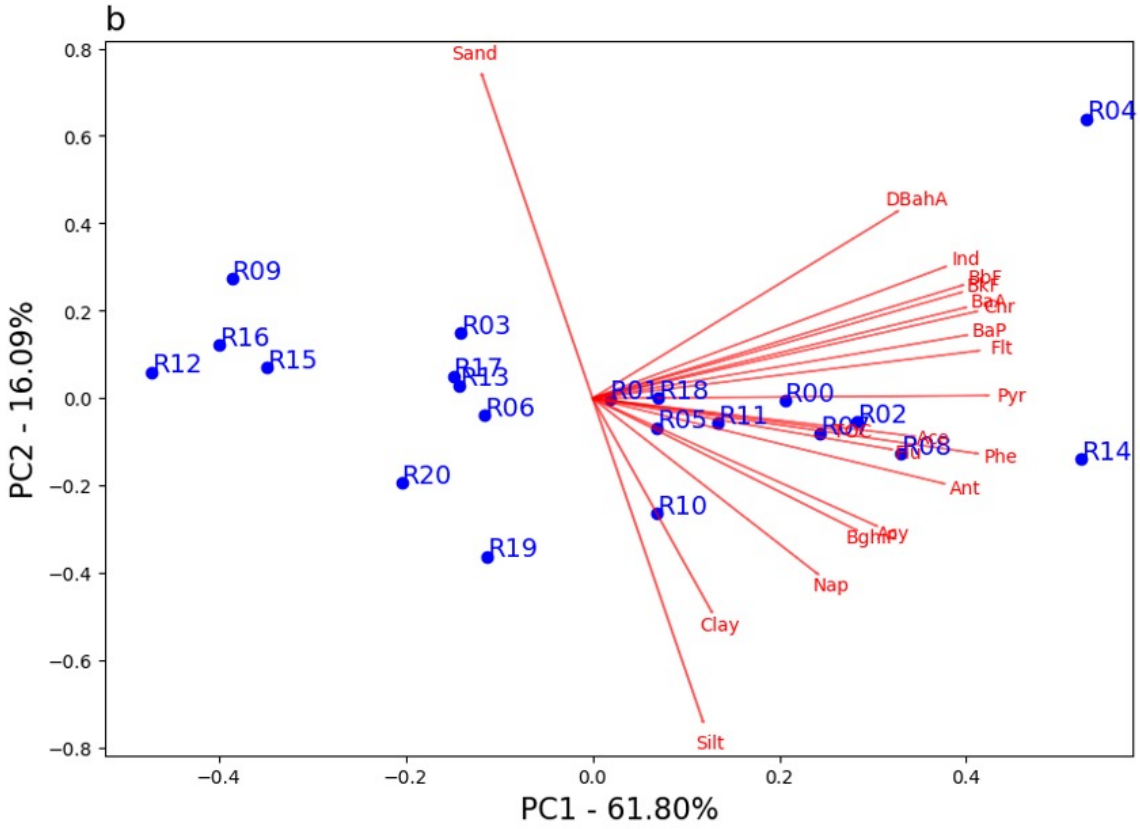
Annex 27: PAH diagnostic ratios determined on the Red River sediment samples.

Stations	Campaigns	BaA/(BaA+Chr)	LMW/HMW	Ind/(Ind+BghiP)	Flt/(Flt+Pyr)
R00	03/2023	0.87	0.37	0.54	0.90
	06/2023	0.36	0.34	0.43	0.52
	09/2023	0.33	0.25	0.42	0.53
R01	03/2023	0.69	0.35	0.48	0.59
	06/2023	0.41	0.28	0.43	0.57
	09/2023	0.58	0.33	0.43	0.52
R02	03/2023	0.60	0.27	0.45	0.60
	06/2023	0.38	0.40	0.39	0.54
	09/2023	0.56	0.25	0.38	0.47
R03	03/2023	0.57	0.31	0.44	0.61
	06/2023	0.37	0.36	0.45	0.55
	09/2023	0.37	0.26	0.44	0.52
R04	03/2023	0.79	0.36	0.52	0.66
	06/2023	0.37	0.28	0.79	0.59
	09/2023	0.54	0.19	0.47	0.58
R05	03/2023	0.58	0.22	0.42	0.59
	06/2023	0.38	0.32	0.42	0.55
	09/2023	0.31	0.27	0.42	0.54
R06	03/2023	0.83	0.26	0.49	0.66
	06/2023	0.40	0.41	0.47	0.55
	09/2023	0.61	0.28	0.40	0.56
R07	03/2023	0.75	0.33	0.50	0.59
	06/2023	0.35	0.42	0.44	0.52
	09/2023	0.61	0.27	0.43	0.54
R08	03/2023	0.73	0.27	0.45	0.62
	06/2023	0.36	0.37	0.41	0.54
	09/2023	0.68	0.22	0.39	0.39
R09	03/2023	0.60	0.27	0.47	0.65
	06/2023	0.41	0.25	0.49	0.57
	09/2023	0.44	0.25	0.42	0.51
R10	03/2023	0.71	0.45	0.58	0.64
	06/2023	0.39	0.54	0.44	0.54
	09/2023	0.63	0.35	0.42	0.56
R11	03/2023	0.76	0.23	0.50	0.57
	06/2023	0.39	0.42	0.41	0.53
	09/2023	0.58	0.31	0.41	0.60
R12	03/2023	0.85	0.22	0.50	0.57
	06/2023	0.42	0.22	0.51	0.57
	09/2023	0.62	0.12	0.54	0.47

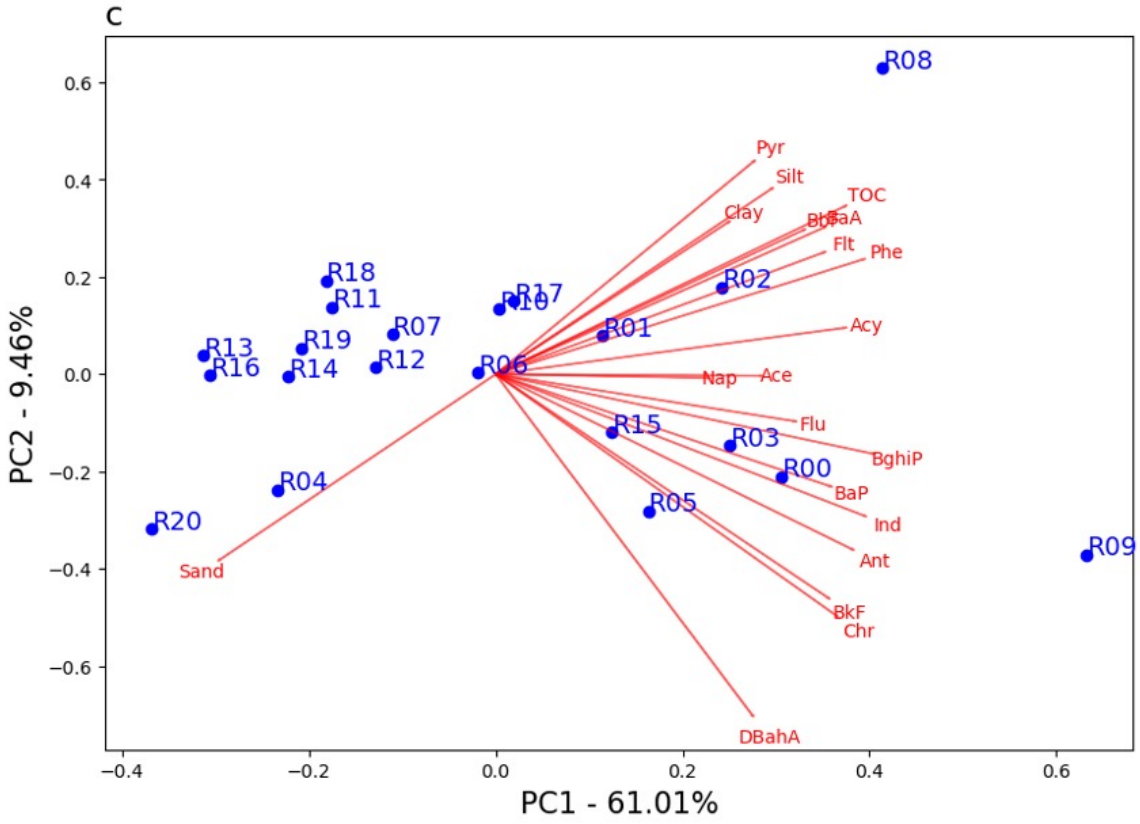
R13	03/2023	0.89	0.26	0.49	0.57
	06/2023	0.41	0.36	0.44	0.54
	09/2023	0.50	0.41	0.50	0.55
R14	03/2023	0.64	0.28	0.49	0.55
	06/2023	0.40	0.38	0.41	0.55
	09/2023	0.56	0.32	0.48	0.52
R15	03/2023	0.75	0.25	0.50	0.60
	06/2023	0.40	0.34	0.46	0.53
	09/2023	0.35	0.36	0.43	0.54
R16	03/2023	1.00	0.31	0.55	0.73
	06/2023	0.42	0.32	0.48	0.56
	09/2023	0.55	0.23	0.49	0.46
R17	03/2023	1.00	0.30	0.55	0.67
	06/2023	0.40	0.26	0.42	0.54
	09/2023	0.60	0.23	0.42	0.57
R18	03/2023	0.75	0.30	0.52	0.59
	06/2023	0.39	0.30	0.44	0.55
	09/2023	0.54	0.36	0.45	0.53
R19	03/2023	0.90	0.36	0.52	0.20
	06/2023	0.35	0.50	0.46	0.55
	09/2023	0.57	0.19	0.49	0.55
R20	03/2023	0.92	0.33	0.55	0.61
	06/2023	0.43	0.50	0.45	0.59
	09/2023	0.42	0.14	0.58	0.41

Annex 28: PCA analysis of 16 PAHs, TOC (%), and sediment size distribution on 21 sediment samples from the Red River in a) March, b) June, and c) September 2023.





Annex


C
R08
Pyr
Silt
TOC
Clay
BbfA
Flt
Phe
R18
R11
R17
R02
R07
R01
Acy
R13
R19
R16
R14
R12
R06
Nap
Ace
Flu
R15
R03
BghiP
R00
R04
BaP
R05
Ind
R20
Ant
R09
Sand
BkF
Chr
DBahA
PC2 - 9.46%
PC1 - 61.01%
0.6
0.4
0.2
0.0
−0.2
−0.4
−0.6
−0.4
−0.2
0.0
0.2
0.4
0.6

Annex

Annex 29: The extracted eigenvectors of PCA for sediment samples in three sampling campaigns

	March 2023		June 2023		September 2023	
	PC1	PC2	PC1	PC2	PC1	PC2
Nap	0.09	0.44	0.16	-0.27	0.15	-0.01
Acy	0.18	0.42	0.20	-0.19	0.25	-0.06
Ace	0.13	0.09	0.23	-0.06	0.18	-0.002
Flu	0.25	0.13	0.21	-0.08	0.21	-0.06
Phe	0.25	0.19	0.27	-0.08	0.26	0.16
Ant	0.21	-0.27	0.25	-0.13	0.25	-0.24
Flt	0.27	0.17	0.27	0.07	0.23	0.17
Pyr	0.28	0.07	0.28	0.004	0.18	0.29
BaA	0.27	-0.20	0.26	0.14	0.23	0.20
Chr	0.27	0.11	0.27	0.13	0.24	-0.33
BbF	0.28	-0.05	0.26	0.17	0.22	0.20
BkF	0.21	0.19	0.26	0.16	0.24	-0.31
BaP	0.28	-0.03	0.26	0.09	0.24	-0.15
DBahA	0.11	-0.45	0.22	0.28	0.18	-0.47
Ind	0.27	-0.08	0.25	0.20	0.26	-0.19
BghiP	0.27	0.001	0.18	-0.20	0.27	-0.11
TOC	0.19	-0.15	0.17	-0.05	0.25	0.23
Clay	0.13	0.001	0.08	-0.33	0.17	0.21
Silt	0.17	-0.25	0.08	-0.49	0.20	0.25
Sand	-0.17	0.25	-0.08	0.49	-0.20	-0.25
Variance (%)	56.54	11.08	61.80	16.09	61.01	9.46

Annex 30: TEQ and MEQ values

Stations	Campaigns	TEQ (ng g^{-1} dw)	MEQ (ng g^{-1} dw)
R00	03/2023	1.93	2.09
	06/2023	6.14	6.88
	09/2023	5.17	6.08
R01	03/2023	3.38	3.83
	06/2023	5.81	6.43
	09/2023	1.91	2.95
R02	03/2023	5.10	6.08
	06/2023	6.94	7.95
	09/2023	4.36	6.21
R03	03/2023	4.50	5.38
	06/2023	4.50	4.85
	09/2023	5.58	6.40
R04	03/2023	2.61	2.84
	06/2023	8.95	10.48
	09/2023	2.56	3.52
R05	03/2023	5.45	6.56
	06/2023	5.76	6.44
	09/2023	4.44	5.12
R06	03/2023	2.98	3.27
	06/2023	4.76	5.07
	09/2023	2.99	3.81
R07	03/2023	3.39	3.77
	06/2023	6.17	6.87
	09/2023	2.20	2.87
R08	03/2023	4.68	5.62
	06/2023	6.71	7.73
	09/2023	4.99	7.30
R09	03/2023	4.34	5.25
	06/2023	3.96	3.99
	09/2023	7.56	9.41
R10	03/2023	2.61	2.82
	06/2023	5.14	5.50
	09/2023	2.34	3.41
R11	03/2023	4.20	4.73
	06/2023	5.77	6.30
	09/2023	1.41	2.45
R12	03/2023	3.69	4.05
	06/2023	3.34	3.38
	09/2023	3.41	4.67

	03/2023	3.01	3.30
R13	06/2023	4.90	5.36
	09/2023	0.76	1.24
	03/2023	3.92	4.66
R14	06/2023	7.11	8.25
	09/2023	1.52	2.06
	03/2023	3.17	3.60
R15	06/2023	3.86	3.88
	09/2023	3.28	3.66
	03/2023	2.95	3.01
R16	06/2023	3.68	3.64
	09/2023	0.93	1.45
	03/2023	3.38	3.40
R17	06/2023	5.09	5.61
	09/2023	3.25	4.97
	03/2023	3.35	3.79
R18	06/2023	5.44	6.09
	09/2023	1.19	1.95
	03/2023	3.13	3.30
R19	06/2023	3.96	4.11
	09/2023	1.76	3.00
	03/2023	2.74	2.88
R20	06/2023	3.64	3.75
	09/2023	2.01	1.81

Annex 31: PCB concentrations (ng g^{-1} dw) in sediment samples collected in November 2022 and June 2023.

November 2022							
	PCB 28	PCB 52	PCB 101	PCB 118	PCB 138	PCB 153	PCB 180
R00	< LOD	0.61	2.55	0.66	13.36	6.29	5.81
R01	< LOD	< LOD	0.41	< LOD	2.61	1.47	1.16
R02	< LOD	0.15	1.03	0.38	6.88	3.65	4.40
R03	< LOD	< LOD	0.30	< LOD	2.44	1.33	1.61
R04	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD
R05	< LOD	< LOD	0.31	< LOD	2.36	1.33	1.59
R06	< LOD	0.26	1.30	0.33	6.70	3.73	3.38
R07	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD
R08	< LOD	< LOD	0.36	< LOD	2.50	1.20	0.99
R09	< LOD	< LOD	0.13	< LOD	0.86	0.47	0.36
R10	< LOD	< LOD	0.14	< LOD	1.10	0.65	0.67
R11	< LOD	< LOD	0.11	< LOD	0.86	0.55	0.58
R12	< LOD	0.70	4.05	1.65	20.09	11.10	12.89
R13	< LOD	0.14	1.26	0.49	8.24	4.32	4.81
R14	< LOD	< LOD	0.17	< LOD	1.47	0.93	1.16
R15	< LOD	< LOD	0.25	< LOD	1.41	0.84	0.91
R16	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD
R17	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD
R18	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD
R19	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD
R20	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD
June 2023							
	PCB 28	PCB 52	PCB 101	PCB 118	PCB 138	PCB 153	PCB 180
R00	< LOD	< LOD	< LOD	0.06	< LOD	< LOD	< LOD
R01	< LOD	< LOD	< LOD	0.07	0.14	< LOD	0.16
R02	< LOD	< LOD	< LOD	0.08	< LOD	< LOD	< LOD
R03	< LOD	< LOD	< LOD	0.15	0.13	< LOD	< LOD
R04	< LOD	< LOD	< LOD	0.14	0.13	< LOD	< LOD
R05	< LOD	< LOD	< LOD	0.11	0.12	< LOD	< LOD
R06	< LOD	< LOD	< LOD	0.08	< LOD	< LOD	< LOD
R07	< LOD	< LOD	< LOD	0.11	0.11	< LOD	< LOD
R08	< LOD	< LOD	< LOD	0.07	< LOD	< LOD	< LOD
R09	< LOD	< LOD	< LOD	0.08	< LOD	< LOD	< LOD
R10	< LOD	< LOD	< LOD	0.10	< LOD	< LOD	< LOD
R11	< LOD	< LOD	< LOD	0.06	< LOD	< LOD	< LOD
R12	< LOD	< LOD	< LOD	0.06	< LOD	< LOD	< LOD
R13	< LOD	< LOD	< LOD	0.07	< LOD	< LOD	< LOD

R14	< LOD	< LOD	< LOD	0.06	< LOD	< LOD	< LOD
R15	< LOD	< LOD	< LOD	0.07	< LOD	< LOD	< LOD
R16	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD
R17	< LOD	< LOD	< LOD	0.06	< LOD	< LOD	< LOD
R18	< LOD	< LOD	< LOD	0.07	< LOD	< LOD	< LOD
R19	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD
R20	< LOD	< LOD	< LOD	0.06	< LOD	< LOD	< LOD

Annex 32 : Spearman correlation for the PCBs detected in sediment samples from two sampling campaigns (p-value < 0.01)

	PCB 28	PCB 52	PCB 101	PCB 118	PCB 138	PCB 153	PCB 180
PCB 28	-						
PCB 52	-	1					
PCB 101	-	0.67	1				
PCB 118	-	0.58	-	1			
PCB 138	-	0.61	0.92	-	1		
PCB 153	-	0.66	0.98	-	0.94	1	
PCB 180	-	0.65	0.97	-	0.93	1	1

"-": signifies non-significant correlations (p-value > 0.01)

Annex 33: RQ values for PCBs in the 21 sediment samples collected along the Red River during dry (November 2022) and rainy (June 2023) seasons.

	November 2022	June 2023
R00	0.1057	0.0002
R01	0.0204	0.0013
R02	0.0595	0.0003
R03	0.0205	0.0010
R04	n.d.	0.0010
R05	0.0202	0.0008
R06	0.0567	0.0003
R07	n.d.	0.0008
R08	0.0182	0.0003
R09	0.0066	0.0003
R10	0.0093	0.0003
R11	0.0076	0.0002
R12	0.1822	0.0002
R13	0.0695	0.0002
R14	0.0135	0.0002
R15	0.0123	0.0002
R16	n.d.	n.d.
R17	n.d.	0.0002
R18	n.d.	0.0004
R19	n.d.	n.d.
R20	n.d.	0.0003

Annex

Annex 34: SCCP concentrations (ng g^{-1} dw and ng g^{-1} normalized to 1% TOC according to Canadian Environmental sediment quality guidelines) in sediment samples collected along the Red River in November 2022 and June 2023.

	November 2022		June 2023	
	ng g^{-1} dw	ng g^{-1} normalized to 1% TOC	ng g^{-1} dw	ng g^{-1} normalized to 1% TOC
R00	270.0	503.5	132.2	169.8
R01	159.5	121.6	131.3	107.3
R02	224.2	668.8	163.6	260.8
R03	116.9	102.0	126.1	146.5
R04	156.7	442.6	388.0	441.2
R05	242.8	346.5	127.8	192.5
R06	230.0	318.0	113.8	176.2
R07	151.8	359.6	211.6	223.6
R08	167.1	138.0	238.1	200.5
R09	178.7	281.2	116.2	254.0
R10	293.6	342.9	77.2	108.4
R11	229.1	282.7	632.8	802.7
R12	166.4	202.2	< LOD	< LOD
R13	< LOD	< LOD	885.5	1456.8
R14	49.1	64.6	1515.7	2144.3
R15	< LOD	< LOD	252.8	511.7
R16	44.0	62.3	1341.3	3247.3
R17	< LOD	< LOD	116.3	154.0
R18	< LOD	< LOD	102.8	112.7
R19	< LOD	< LOD	< LOD	< LOD
R20	< LOD	< LOD	634.9	1143.3

Annex

Annex 35: List of analyzed sediment and water samples for PFAS.

	November 2022		March 2023		June 2023		September 2023	
Stations	Water	Sediment	Water	Sediment	Water	Sediment	Water	Sediment
R00		✓	✓	✓		✓		✓
R01		✓		✓	✓	✓		✓
R02		✓	✓	✓	✓	✓		✓
R03		✓	✓	✓	✓	✓		✓
R04	✓	✓	✓	✓		✓		✓
R06	✓	✓	✓	✓	✓	✓		✓
R09	✓	✓	✓	✓	✓	✓		✓
R10	✓	✓						
R13	✓	✓						
R14	✓	✓		✓	✓	✓		✓
R15		✓	✓	✓		✓		✓
R18		✓	✓	✓	✓	✓		✓
R20		✓	✓	✓	✓	✓		✓

Annex

Annex 36: Concentration of PFAS (ng L^{-1} and ng F L^{-1} in bracket) detected by target approach in water sample from November 2022

PFAS	R04	R06	R09	R10	R13	R14
PFHxA	n.d.	n.d.	n.d.	n.d.	n.d.	0.98 (0.65)
PFHpA	0.11 (0.07)	n.d.	n.d.	0.09 (0.06)	0.09 (0.06)	0.45 (0.31)
PFOA	0.31 (0.21)	0.17 (0.12)	0.23 (0.16)	0.39 (0.27)	0.39 (0.27)	0.62 (0.42)
HPFO-DA	< LOD	< LOD	n.d.	< LOD	< LOD	4.95 (3.13)
4:2-FTS	n.d.	1.46 (0.76)	n.d.	n.d.	n.d.	5.83 (3.05)
6:2-FTS	n.d.	203.67 (117.49)	28.99 (16.72)	8.51 (4.91)	1.35 (0.78)	557.05 (321.35)
8:2-FTS	n.d.	n.d.	n.d.	n.d.	n.d.	1.62 (0.99)
6:2-diPAP	n.d.	1.09 (0.68)	n.d.	n.d.	< LOD	< LOD
∑PFAS	0.42 (0.28)	206.39 (119.05)	29.22 (16.88)	8.99 (5.24)	1.83 (1.11)	571.5 (329.9)

Annex

Annex 37: Concentration of PFAS (ng L^{-1} and ng F L^{-1} in bracket) detected by target approach in water sample from March 2023.

PFAS	R00	R02	R03	R04	R06	R09	R15	R18	R20
PFBA	n.d.	n.d.	n.d.	7.67 (4.76)	n.d.	n.d.	n.d.	n.d.	n.d.
PFPeA	n.d.	n.d.	n.d.	0.99 (0.64)	n.d.	n.d.	n.d.	n.d.	n.d.
PFHxA	n.d.	n.d.	n.d.	1.28 (0.85)	n.d.	n.d.	n.d.	n.d.	0.49 (0.33)
PFHpA	0.06 (0.04)	0.08 (0.05)	n.d.	0.45 (0.31)	0.06 (0.04)	0.07 (0.04)	0.1 (0.06)	0.05 (0.03)	0.04 (0.03)
PFOA	0.14 (0.1)	0.12 (0.08)	0.19 (0.13)	0.59 (0.41)	0.18 (0.13)	0.17 (0.12)	0.17 (0.12)	0.18 (0.12)	0.29 (0.2)
PFBS	n.d.	n.d.	n.d.	n.d.	2.59 (1.47)	1.92 (1.1)	n.d.	n.d.	n.d.
∑PFAS	0.2 (0.14)	0.2 (0.13)	0.19 (0.13)	10.98 (6.97)	2.83 (1.64)	2.16 (1.26)	0.27 (0.18)	0.23 (0.15)	0.82 (0.56)

Annex 38: Concentration of PFAS (ng L^{-1} and ng F L^{-1} in bracket) detected by target approach in water sample from June 2023.

PFAS	R01	R02	R03	R06	R09	R14	R18	R20

PFBA	1.14 (0.71)	1.3 (0.81)	1.12 (0.7)	n.d.	n.d.	n.d.	n.d.	n.d.
PFPeA	n.d.	0.27 (0.18)	0.24 (0.16)	n.d.	n.d.	n.d.	n.d.	n.d.
PFHpA	0.14 (0.1)	0.18 (0.12)	0.2 (0.13)	0.16 (0.11)	< LOD	< LOD	< LOD	0.14 (0.1)
PFOA	0.27 (0.18)	0.24 (0.17)	0.28 (0.19)	0.29 (0.2)	0.23 (0.16)	0.2 (0.14)	0.25 (0.17)	0.31 (0.21)
HPFO-DA	0.55 (0.35)	0.61 (0.38)	0.63 (0.4)	0.69 (0.44)	< LOD	< LOD	< LOD	0.4 (0.25)
6:2-FTS	n.d.	n.d.	n.d.	0.64 (0.37)	n.d.	n.d.	n.d.	n.d.
FOSAA	0.76 (0.44)	< LOD	< LOD	< LOD	n.d.	n.d.	n.d.	n.d.
N-EtFOSAA	0.06 (0.03)	0.05 (0.03)	0.16 (0.09)	0.14 (0.08)	n.d.	n.d.	n.d.	n.d.
6:2-diPAP	1.24 (0.77)	0.78 (0.49)	0.54 (0.34)	< LOD	< LOD	< LOD	< LOD	n.d.
8:2-diPAP	1.21 (0.79)	< LOD	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
∑PFAS	5.37	3.43	3.17	1.92	0.23	0.2	0.25	0.85

	(3.37)	(2.18)	(2.01)	(1.2)	(0.16)	(0.14)	(0.17)	(0.56)

Annex 39: Welch's t-test result for the comparison of the mean PFAS between November 2022 and March 2023 water samples.

	November 2023	*March 2023*
Mean	136.391409	1.98545375
Variance	51706.0446	12.3154572
Observations	6	9
Hypothesized Mean Difference	0	
df	5	
t Stat	1.44773476	
P(T<=t) one-tail	**0.10367453**	
t Critical one-tail	2.01504837	
P(T<=t) two-tail	0.20734906	
t Critical two-tail	2.57058184	

Annex 40: Welch's t-test result for the comparison of the mean PFAS between November 2022 and June 2023 water samples.

	November 2023	*June 2023*
Mean	136.391409	1.92727675
Variance	51706.0446	3.63344776
Observations	6	8
Hypothesized Mean Difference	0	
df	5	
t Stat	1.44843822	
P(T<=t) one-tail	**0.10358115**	
t Critical one-tail	2.01504837	
P(T<=t) two-tail	0.20716229	
t Critical two-tail	2.57058184	

Annex 41: Welch's t-test result for the comparison of the mean PFAS between March 2023 and June 2023 water samples.

	March 2023	*June 2023*
Mean	1.98545375	1.92727675
Variance	12.3154572	3.63344776
Observations	9	8
Hypothesized Mean Difference	0	
df	13	
t Stat	0.04309331	
P(T<=t) one-tail	**0.48314101**	
t Critical one-tail	1.7709334	
P(T<=t) two-tail	0.96628202	
t Critical two-tail	2.16036866	

Annex 42: PCA loading scores of PFAS in the Red River water samples.

	PC1	PC2
PFBA	0.40	0.88
PFPeA	0.40	0.83
PFHxA	0.86	0.34
PFHpA	0.89	0.37
PFOA	0.87	0.26
PFBS	-0.17	-0.15
PFHxS	0.00	0.00
FBSA	0.00	0.00
HPFO-DA	0.89	-0.36
4:2-FTS	0.87	-0.47
6:2-FTS	0.85	-0.47
8:2-FTS	0.90	-0.45
FOSAA	-0.03	0.38
N-EtFOSAA	0.00	0.31
6:2-diPAP	-0.02	0.33
8:2-diPAP	-0.03	0.38

Annex 43: Concentration of PFAS (ng.L^{-1} and ng F.L^{-1} in bracket) detected by TOPA approach in sediment samples from November 2022

PFAS	R00	R01	R02	R03	R04	R06	R09	R10	R13	R14	R15	R18	R20
PFPeA	0.51 (0.33)	n.d.	n.d.	0.73 (0.47)	1.35 (0.88)	1.16 (0.75)	1.61 (1.04)	n.d.	0.71 (0.46)	1.12 (0.73)	0.98 (0.63)	0.75 (0.49)	0.45 (0.29)
PFHxA	n.d.	n.d.	n.d.	1.12 (0.75)	1.62 (1.08)	1.15 (0.76)	1.72 (1.15)	n.d.	n.d.	1.33 (0.89)	1.26 (0.84)	n.d.	n.d.
PFHpA	n.d.	n.d.	n.d.	0.52 (0.35)	1.37 (0.93)	0.5 (0.34)	1.33 (0.9)	n.d.	n.d.	0.77 (0.53)	0.78 (0.53)	0.32 (0.22)	0.46 (0.31)
PFOA	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	1.24 (0.86)	n.d.	n.d.	0.98 (0.68)	1.33 (0.92)	n.d.	< LOD
PFNA	n.d.	n.d.	n.d.	0.25 (0.17)	0.16 (0.11)	n.d.	0.46 (0.32)	n.d.	n.d.	0.29 (0.2)	0.31 (0.21)	< LOD	< LOD
PFDA	n.d.	n.d.	n.d.	0.3 (0.21)	0.13 (0.09)	n.d.	0.35 (0.24)	n.d.	n.d.	< LOD	0.23 (0.16)	0.14(0.1)	n.d.
PFUdA	n.d.	n.d.	n.d.	0.65 (0.46)	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
PFDoA	n.d.	n.d.	< LOD	0.96 (0.69)	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.

PFTrDA	n.d.	n.d.	0.51 (0.37)	0.82 (0.59)	n.d.	< LOD	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
PFNS	n.d.	n.d.	n.d.	0.38 (0.25)	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
PFDS	n.d.	n.d.	< LOD	0.69 (0.46)	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
PFUnS	0.3 (0.2)	n.d.	3.15 (2.12)	5.07 (3.41)	n.d.	< LOD	n.d.	n.d.	n.d.	n.d.	n.d.	0.69 (0.46)	0.19(0.13)
PFDoS	n.d.	n.d.	< LOD	0.27 (0.18)	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
∑PFAS	0.81 (0.53)	n.d.	3.66 (2.49)	11.76 (7.99)	4.63 (3.09)	2.81 (1.85)	6.71 (4.51)	n.d.	0.71(0.46)	4.49 (3.03)	4.89 (3.29)	1.9 (1.27)	1.1 (0.73)

Annex 44: Concentration of PFAS (ng.L^{-1} and ng F.L^{-1} in bracket) detected by TOPA approach in sediment samples from March 2023

PFAS	R00	R01	R02	R03	R04	R06	R09	R14	R15	R18	R20
PFPrA	< LOD	3.54 (2.05)	n.d.	3.3 (1.91)	< LOD	3.83 (2.22)	n.d.	n.d.	2.55 (1.48)	n.d.	4.8 (2.78)
PFPeA	n.d.	0.78	n.d.	n.d.	1.42	1.7	n.d.	1.39	0.78	n.d.	n.d.

		(0.51)			(0.92)	(1.1)		(0.9)	(0.5)		
PFHxA	n.d.	1.09 (0.73)	n.d.	0.93 (0.62)	1.39 (0.93)	1.21 (0.8)	n.d.	1.68 (1.12)	0.88 (0.59)	0.79 (0.53)	n.d.
PFHpA	0.3 (0.2)	0.67 (0.45)	0.4 (0.27)	1.46 (0.99)	1.22 (0.83)	0.89 (0.61)	n.d.	0.9 (0.61)	0.84 (0.57)	0.54 (0.36)	0.38 (0.26)
PFOA	n.d.	n.d.	n.d.	1.98 (1.37)	n.d.	1.28 (0.88)	n.d.	n.d.	n.d.	n.d.	n.d.
PFNA	n.d.	n.d.	n.d.	0.5 (0.35)	n.d.	0.29 (0.2)	n.d.	0.22 (0.16)	n.d.	n.d.	n.d.
PFDA	n.d.	< LOD	n.d.	0.45 (0.31)	n.d.	0.33 (0.23)	n.d.	0.35 (0.24)	< LOD	0.25 (0.18)	0.15 (0.1)
PFOS	0.43 (0.28)	n.d.	0.29 (0.19)	0.4 (0.26)	0.27 (0.17)	n.d.	n.d.	n.d.	0.26 (0.17)	0.33 (0.22)	0.36 (0.23)
∑PFAS	0.73(0.48)	6.08(3.74)	0.69 (0.46)	9.02 (5.81)	4.3 (2.85)	9.53 (6.04)	n.d.	4.54 (3.03)	5.31 (3.31)	1.91 (1.29)	5.69 (3.37)

Annex 45: Concentration of PFAS (ng.L^{-1} and ng F.L^{-1} in bracket) detected by TOPA approach in sediment samples from June 2023

PFAS	R00	R01	R02	R03	R04	R06	R09	R14	R15	R18	R20
PFBA	n.d.	1.8 (1.12)	n.d.	n.d.	1.41 (0.88)	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
PFPeA	0.73 (0.47)	0.99 (0.64)	1.2 (0.78)	1.3 (0.84)	2.26 (1.46)	0.92 (0.59)	0.68 (0.44)	n.d.	0.77 (0.5)	n.d.	n.d.
PFHxA	n.d.	1.03 (0.69)	1.35 (0.9)	1.74 (1.16)	3.04(2.02)	1.05 (0.7)	0.82 (0.55)	n.d.	0.99 (0.66)	n.d.	n.d.
PFHpA	0.52 (0.35)	0.44 (0.3)	0.75 (0.51)	1.44 (0.98)	3.16 (2.14)	0.83 (0.56)	0.65 (0.44)	0.96 (0.65)	0.69 (0.47)	0.17 (0.12)	0.35 (0.24)
PFOA	n.d.	n.d.	1.22 (0.84)	2.01 (1.38)	2.72 (1.87)	1.25 (0.86)	0.92 (0.63)	1.14 (0.78)	1.04 (0.71)	n.d.	n.d.
PFNA	< LOD	< LOD	0.37 (0.26)	0.72 (0.5)	1.09 (0.76)	0.49 (0.34)	0.33 (0.23)	0.34 (0.23)	0.26 (0.18)	n.d.	0.2 (0.14)
PFDA	n.d.	0.18 (0.12)	0.29 (0.2)	0.56 (0.39)	0.68 (0.48)	0.32 (0.22)	0.17 (0.12)	0.32 (0.23)	0.23 (0.16)	< LOD	0.19 (0.13)

PFOS	n.d.	0.23 (0.15)	0.4 (0.26)	0.21 (0.14)	0.23 (0.15)	0.35 (0.23)	0.27 (0.18)	0.22 (0.14)	0.21 (0.13)	n.d.	0.28 (0.18)
∑PFAS	1.25 (0.82)	4.67 (3.02)	5.58 (3.75)	7.98 (5.39)	14.59 (9.76)	5.21 (3.5)	3.84 (2.59)	2.98 (2.03)	4.19 (2.81)	0.17 (0.12)	1.02 (0.69)

Annex 46: Concentration of PFAS (ng L^{-1} and ng F L^{-1} in bracket) detected by TOPA approach in sediment samples from September 2023

PFAS	R00	R01	R02	R03	R04	R06	R09	R14	R15	R18	R20
PFPeA	n.d.	n.d.	0.6 (0.39)	0.78 (0.51)	0.72 (0.46)	0.59 (0.38)	1.45 (0.94)	0.54 (0.35)	n.d.	0.59 (0.38)	n.d.
PFHxA	n.d.	n.d.	n.d.	1.2 (0.8)	0.78 (0.52)	0.7 (0.47)	1.65 (1.1)	0.68 (0.45)	n.d.	0.67 (0.44)	n.d.
PFHpA	0.24 (0.16)	0.25 (0.17)	0.36 (0.25)	1.01(0.69)	0.67 (0.46)	0.54 (0.37)	1.29 (0.87)	0.38 (0.26)	0.44 (0.3)	0.63 (0.43)	n.d.
PFOA	n.d.	n.d.	n.d.	1.63 (1.12)	< LOD	n.d.	1.64 (1.13)	0(0)	< LOD	< LOD	n.d.
PFNA	< LOD	0.27 (0.19)	0.22 (0.15)	0.57 (0.4)	< LOD	0.2 (0.14)	0.32 (0.22)	0.03 (0.02)	0.26 (0.18)	0.3 (0.21)	< LOD

PFDA	< LOD	< LOD	< LOD	0.25 (0.17)	< LOD	< LOD	0.37(0.26)	0(0)	0.14 (0.1)	0.18 (0.12)	< LOD
∑PFAS	0.24 (0.16)	0.52 (0.36)	1.18 (0.79)	5.44 (3.69)	2.17 (1.44)	2.03 (1.36)	6.72 (4.52)	1.63 (1.08)	0.84 (0.58)	2.37 (1.58)	n.d.

Annex 47: Concentration of PFAS (ng L^{-1} and ng F L^{-1} in bracket) detected by TOPA approach in water samples from November 2022

PFAS	R04	R06	R09	R10	R13	R14
PFPrA	10.45 (6.05)	26.01 (15.06)	25.09 (14.53)	21.46 (12.43)	19.15 (11.09)	35.58 (20.61)
PFBA	2.61 (1.62)	17.21 (10.69)	5.15 (3.2)	2.31 (1.43)	2.91 (1.81)	55.88 (34.72)
PFPeA	1.38 (0.89)	29.52 (19.12)	5.92 (3.84)	2.16 (1.4)	0.62 (0.4)	110.43 (71.52)
PFHxA	0.7 (0.46)	35.26 (23.46)	6.22 (4.14)	1.72 (1.14)	0.19 (0.13)	133.56 (88.88)
PFHpA	1.07 (0.73)	3.32 (2.25)	1.03 (0.7)	2.49 (1.69)	1.33 (0.9)	13.7 (9.3)
PFOA	0.88	0.45	0.63	0.35	0.68	0.7

	(0.61)	(0.31)	(0.43)	(0.24)	(0.47)	(0.48)
PFDA	< LOD	n.d.	< LOD	< LOD	n.d.	0.14 (0.1)
∑PFAS	17.09 (10.36)	111.77 (70.89)	44.04 (26.84)	30.49 (18.33)	24.88 (14.8)	349.99 (225.61)

Annex 48: Concentration of PFAS (ng L^{-1} and ng F L^{-1} in bracket) detected by TOPA approach in water samples from March 2023

PFAS	R00	R02	R03	R04	R06	R09	R15	R18	R20
PFPrA	3.1 (1.79)	< LOD	4.91 (2.84)	< LOD	< LOD	2.1 (1.21)	2.03 (1.17)	7.43 (4.3)	n.d.
PFBA	2.12 (1.32)	1.71 (1.06)	2.85 (1.77)	1.35 (0.84)	2.28 (1.42)	1.64 (1.02)	1.39 (0.87)	3.6 (2.24)	1.34 (0.83)
PFPeA	0.53 (0.35)	0.23 (0.15)	1.7 (1.1)	0.91 (0.59)	0.27 (0.17)	0.33 (0.21)	0.43(0.28)	0.77 (0.5)	0.11 (0.07)
PFHxA	0.31 (0.21)	0.1 (0.07)	0.57 (0.38)	1.01 (0.67)	0.12 (0.08)	0.19 (0.12)	0.28(0.19)	0.56 (0.37)	0.25 (0.17)
PFHpA	n.d.	n.d.	0.03 (0.02)	n.d.	n.d.	n.d.	n.d.	0.11 (0.07)	n.d.
PFOA	n.d.	n.d.	0.04	0.21	< LOD	0.16	n.d.	0.1	n.d.

			(0.03)	(0.15)		(0.11)		(0.07)	
PFNA	0.04 (0.02)	n.d.	0.09 (0.06)	< LOD	n.d.	n.d.	n.d.	n.d.	n.d.
PFBS	0.48 (0.27)	0.48 (0.27)	< LOD	< LOD	< LOD	0.49 (0.28)	< LOD	0.62 (0.35)	< LOD
PFHxS	n.d.	n.d.	n.d.	0.02 (0.01)	n.d.	< LOD	0(0)	n.d.	0.02 (0.01)
PFOS	< LOD	n.d.	0.05 (0.04)	0.09 (0.06)	< LOD	< LOD	< LOD	n.d.	< LOD
PFUnS	n.d.	n.d.	n.d.	0.09 (0.06)	n.d.	n.d.	n.d.	n.d.	n.d.
∑PFAS	6.58 (3.96)	2.52 (1.55)	10.24 (6.24)	3.68 (2.38)	2.67 (1.67)	4.91 (2.95)	4.13 (2.51)	13.19 (7.9)	1.72 (1.08)

Annex 49: Concentration of PFAS (ng L^{-1} and ng F L^{-1} in bracket) detected by TOPA approach in water samples from June 2023.

PFAS	R01	R02	R03	R06	R09	R14	R18	R20
PFPrA	14.44 (8.36)	8.57 (4.96)	1.98 (1.15)	2.77 (1.61)	4.45 (2.58)	3.15 (1.82)	< LOD	1.33 (0.77)
PFBA	1.54 (0.96)	4.01 (2.49)	2.33 (1.45)	1.86 (1.15)	1.16 (0.72)	n.d.	1.14 (0.71)	< LOD
PFPeA	0.73 (0.47)	0.49 (0.32)	0.42 (0.27)	0.27 (0.17)	1.42 (0.92)	0.14 (0.09)	1.05 (0.68)	0.17 (0.11)
PFHxA	0.41 (0.27)	0.31 (0.21)	0.29 (0.19)	0.24 (0.16)	0.42 (0.28)	0.04 (0.03)	0.49 (0.33)	0.13 (0.09)
PFHpA	0.73 (0.49)	n.d.	0.14 (0.1)	0.13 (0.09)	0.09 (0.06)	n.d.	n.d.	n.d.
PFOA	0.61 (0.42)	n.d.	n.d.	n.d.	0.68 (0.47)	n.d.	< LOD	n.d.
PFNA	n.d.	n.d.	0.06 (0.04)	0.06 (0.04)	0.2 (0.14)	< LOD	< LOD	< LOD
PFDA	n.d.	n.d.	n.d.	n.d.	LOD()	n.d.	n.d.	n.d.
PFHxS	n.d.	< LOD	< LOD	< LOD	0.02 (0.01)	n.d.	0.04 (0.02)	< LOD

PFOS	n.d.	n.d.	n.d.	< LOD	0.12 (0.08)	< LOD	< LOD	< LOD
∑PFAS	18.46 (10.97)	13.38 (7.98)	5.22 (3.2)	5.33 (3.22)	8.56 (5.26)	3.33 (1.94)	2.72 (1.74)	1.63 (0.97)

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