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# Spatial distribution and source apportionment of polycyclic aromatic hydrocarbons (PAHs) in surficial sediments along the coast of British Columbia, Canada

Elizabeth J. Ruberg [a], Mark B. Yunker [b], Gabriela Aguirre-Martinez [c], Kelsey Delisle [d], Carmen Morales-Caselles [e], Peter S. Ross [f], Marie Noël [g,*]

[a] *Ocean Wise Conservation Association, 440 Cambie Street, Vancouver, British Columbia, Canada, V6B 2N5*
[b] *7137 Wallace Dr., Brentwood Bay, British Columbia, Canada, V8M 1G9*
[c] *Faculty of Health Sciences, Arturo Prat University, Casilla 121, 1110939, Iquique, Chile*
[d] *Fisheries and Oceans Canada, 401 Burrard Street, Vancouver, British Columbia, Canada, V6C 3S4*
[e] *Faculty of Marine and Environmental Sciences, University of Cádiz 11510, Puerto Real, Cádiz, Spain*
[f] *Raincoast Conservation Foundation, 2238 Harbour Rd, Sidney, British Columbia, Canada, V8L 2P6*
[g] *Fisheries and Oceans Canada, 4160 Marine Drive, West Vancouver, British Columbia, Canada, V7V 1H2*

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## ABSTRACT

The 29,000 km coastline of British Columbia (B.C.), Canada, is an area with increasing industrial and shipping activity. Pollution Tracker is the first coast-wide marine pollution monitoring program, generating high resolution contaminant data for sediment samples. Surficial sediment samples were collected as part of the Pollution Tracker Program from 51 sites between 2015 and 2017. Polycyclic aromatic hydrocarbons (PAHs) were detected at all sites, with total PAHs (75 analytes) ranging from 36 to 162,000 nanograms (ng) per gram (g) dry weight (dw). Sediment samples from Prince Rupert Harbour (162,000 ng/g dw) and Victoria Harbour (21,000–47,000 ng/g dw) had the highest PAH levels, consistent with heavy vessel traffic, and a history of urban and marine activities in those areas. Of the 51 sites, 25 % exceeded Canadian marine interim sediment quality guidelines for all 13 PAH analytes. Mean probable effects level quotient values (PEL-Q) indicated 69 % of sites had potential ecological risk associated with PAH exposure from sediment, 25 % of sites had moderate risk, and two sites were categorized as having a high risk of biological effects. Source ratios of four ring and larger parent PAHs indicated combustion is the primary contributor to the hydrocarbon sediment profile, with most samples containing mixed biomass/solid fuel and liquid fuel combustion sources. Correspondingly, petrogenic source apportionment using alkyl naphthalene isomers suggested most samples contained PAHs from both coal and oil sources. Our high-resolution baseline data provides further insight into the sources and fate of PAHs in coastal B.C. marine sediment.

## 1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are ubiquitous in the environment and consist of thousands of compounds originating from both anthropogenic activities and natural sources. Pyrogenic PAHs are formed through the incomplete combustion of organic matter (forest fires, agricultural burning, campfires, and emissions from vehicles and industrial operations), resulting primarily in the formation of parent PAHs. In contrast, petrogenic PAHs are found in fossil fuels (coal, oil, and refined fuels such as diesel and gasoline) and contain the parent PAHs plus a wide range of PAHs with alkylated groups attached to the parent that can be more persistent than their parent. Because of their physico-chemical properties, PAHs are persistent, can be transported over great distances through atmospheric and oceanic currents and can become bioavailable for uptake by organisms (Latimer and Zheng, 2003; Burgess et al., 2003; Meador, 2003; Yang et al., 2022).

In the marine environment, direct natural sources of PAHs include crude oil seeps and coal and shale deposits, while anthropogenic sources include oil spills, oil discharges from vessels and combustion of fuels. Indirect sources to the marine environment include forest fires,

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* Corresponding author.
*E-mail address:* Marie.Noel@dfo-mpo.gc.ca (M. Noël).

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agricultural burning, campfires, industrial activities, and vehicle use. These activities may cause PAHs to enter the marine environment through atmospheric deposition, urban run-off, and discharge from wastewater treatment facilities (Latimer and Zheng, 2003). Forest fires, home firewood burning, coal combustion, vehicle use, and industrial point sources are the largest contributors to PAH emissions in Canada, with forest fires being the top contributor to overall Canada-wide emissions (Berthiaume et al., 2021; Marvin et al., 2021). In British Columbia (B.C.), the use of carbon electrodes at aluminum smelters are major industrial point source contributors of PAH emissions (Marvin et al., 2021).

Distinguished by its beauty, natural resources, and biodiversity (Morales-Caselles et al., 2017a) the coastal environment of B.C., Canada, is home to a rich diversity of species considered invaluable to First Nations communities (Thompson et al., 2017; Marushka et al., 2019, 2021; Andrade-Rivas et al., 2022), commercial and recreational fishers, and ecotourism operators. Thirty-one marine mammal species inhabit the coastal waters of B.C. (Ford, 2014; Alava, 2019) including the Southern Resident killer whale (SRKW) (*Orcinus orca*) population listed as *Endangered* and the Northern Resident killer whale (NRKW) population listed as *Threatened* under the Canadian *Species at Risk Act* (Fisheries and Oceans Canada (DFO), 2018). At the same time, B.C. is under increasing anthropogenic pressure with the presence of major urban and industrial centres, ports, mills, refineries, on-going oil pipeline development and ever-increasing oil tanker traffic leading to major sources of pollution to the marine environment (Alava and Calle, 2017; Alava, 2019). PAHs have been detected in B.C. marine coastal sediment (Yunker et al., 2002; Yunker et al., 2011a; Yunker et al., 2014; Yunker et al., 2015; Yunker and Macdonald, 2003; Harris et al., 2011a and a variety of invertebrate species (Yunker et al., 2011a; Andrade-Rivas et al., 2022; Willie et al., 2017; Thompson et al., 2017; King et al., 2023; Eickhoff et al., 2003). PAHs have also been measured in marine vertebrates on the B.C. coast, such as sea otters (*Enhydra lutris*) (Harris et al., 2011b) and Southern Resident and Bigg's killer whales (Lee et al., 2023).

While parent PAHs can be metabolized by aquatic vertebrates (Collier et al., 2013), alkyl PAHs may biomagnify in vertebrates depending on the metabolic efficiency of their prey species (Harris et al., 2011b). PAHs such as those associated with liquid fossil fuel combustion and petroleum discharges are expected to be fully bioavailable to marine fauna and some of these PAHs are known carcinogenic and mutagenic compounds (Yunker et al., 2015). Exposure to oil spill related PAHs has been associated with killer whale mortalities (Matkin et al., 2008; Franker, 2013) and deleterious health effects in other marine mammals as reviewed in Ruberg et al. (2021) such as adrenal dysfunction (Venn-Watson et al., 2015) and lung disease (Schwacke et al., 2014) in bottlenose dolphins (*Tursiops truncatus*). Chronic exposure to PAHs from aluminum smelter waste has been associated with an increased rate of DNA adducts and gastrointestinal cancers in a population of beluga whales (*Delphinapterus leucas*), (Poirier et al., 2019, 2021; Poirier, 2021; Lair et al., 2016). Additionally, exposure to PAHs adversely affects juvenile salmon (*Oncorhynchus* sp.) (Heintz et al., 2000; Meador et al., 2006), altering both growth and metabolism (Meador et al., 2006; Lundin et al., 2021).

Pollution Tracker was established as a long-term monitoring program (https://pollutiontracker.org) to evaluate contaminant profiles in marine sediment along the B.C. coast. Sediment may act as a sink for PAHs (Harris et al., 2011b; Yunker and Macdonald, 2003; Long et al., 2005), providing a historical record of contamination (Alava, 2019), or as a source of PAHs through remobilization into the water column during natural storms, maintenance dredging (Usanase et al., 2021), and bioturbation (Meador, 2003).The present paper reports PAH levels measured in 51 marine sediment samples collected along the B.C. coast as part of Pollution Tracker Phase 1 (2015–2017), and builds on the results of Yunker et al. (2015). Our results include the evaluation of patterns and 'fingerprinting' to characterize levels and sources of PAHs.

## 2. Materials and methods

### 2.1. Sample collection

Surficial marine sediment samples were collected from 51 sites along the coast of B.C. between January 2015 and February 2017 (Fig. 1, Table A.1). Apart from the site at Saturna Island (SAT), where sediment was collected by a SCUBA diver, nearshore sediment samples were collected using a stainless steel Petite Ponar, Smyth McIntyre or Van Veen grab sampler. Three grabs per location were collected at depths between 1 and 83 m. Average sampling depth was 17.4 m across 49 sites. In addition, two deep samples were collected from the Strait of Georgia (LG63 and LG64) at a depth of 193 and 255 m, respectively. Depositional areas with fine-grained sediment were targeted for sampling. From each grab, the top 2 to 5 cm was collected and combined immediately in a hexane-rinsed stainless steel bowl to create a composite sample for each site. Sediment was maintained in 250-mL amber glass jars at 4 °C prior to arrival at the laboratory, and then frozen at −20 °C for future analysis.

### 2.2. Polycyclic aromatic compounds analysis

Sediment samples were analysed for the major parent PAHs and a broad range of alkylated PAHs (for a total of 75 PAHs) at SGS AXYS Analytical Services Ltd. (Sidney, B.C, Canada) according to SGS AXYS MLA-021. Method MLA-021 is a multi-matrix extended version of EPA 8270C/D/E modified by EPA 1625B (Semi volatile Organic Compounds by Isotope Dilution Gas Chromatography/Mass Spectrometry (GC/MS)) where concentrations of each compound of interest are calculated using isotopic dilution quantification Sediment samples were spiked with deuterated surrogate standards ($n = 18$), then extracted using Soxhlet extraction with dichloromethane. Samples were analysed alongside an ongoing precision and recovery spiked matrix, a reference sample analysed to show continuing conformance to method performance specifications. The extracts were cleaned up with column chromatography on silica, gel permeation chromatography, and treatment with activated copper. This reliable and validated separation through fractionation of the silica column cleanup extract separates alkanes from the PAH fraction, thereby removing a significant source of background matrix (such as the Unresolved Complex Matrix) helping to lower detection limits. Extracts were further cleaned up as necessary by washing with base, additional gel permeation chromatography, or column chromatography on alumina.

Extracts were then analysed by low-resolution mass spectrometry (LRMS) using an RTX-5 capillary GC column. Consistent instrumentation (Agilent 5973 and 5975) with rigid EI control was used for all alkylated PAH analysis. Initial calibration was performed using a five-point calibration series of solutions that encompassed the working concentration range. Initial calibration solutions contained the suite of labelled surrogate and recovery standards and authentic target PAHs, and calibration was verified at least once every 12 h. Alkylated groups were calculated from sums of response for specific ions representing a group over a set period of retention time in one channel. The response of specific, fully quantitative isotope diluted alkylated compounds may occur within this window and it's corresponding second channel. Where this occurs, the value of the section in both channels representing a specific alkylated compound is quantified from the specific compound report with ratioing acceptance for multiple ions criteria applied. This creates responses within alkylated groups that are fully quantitative for many values, compared to open scan semi-quantitative alkylated group "fingerprints" generally used in the industry for alkylated group values.

### 2.3. Additional chromatogram analysis

Peak areas from GC–MS ion traces acquired by SGS AXYS Analytical and the same HP ChemStation software used by SGS AXYS were then

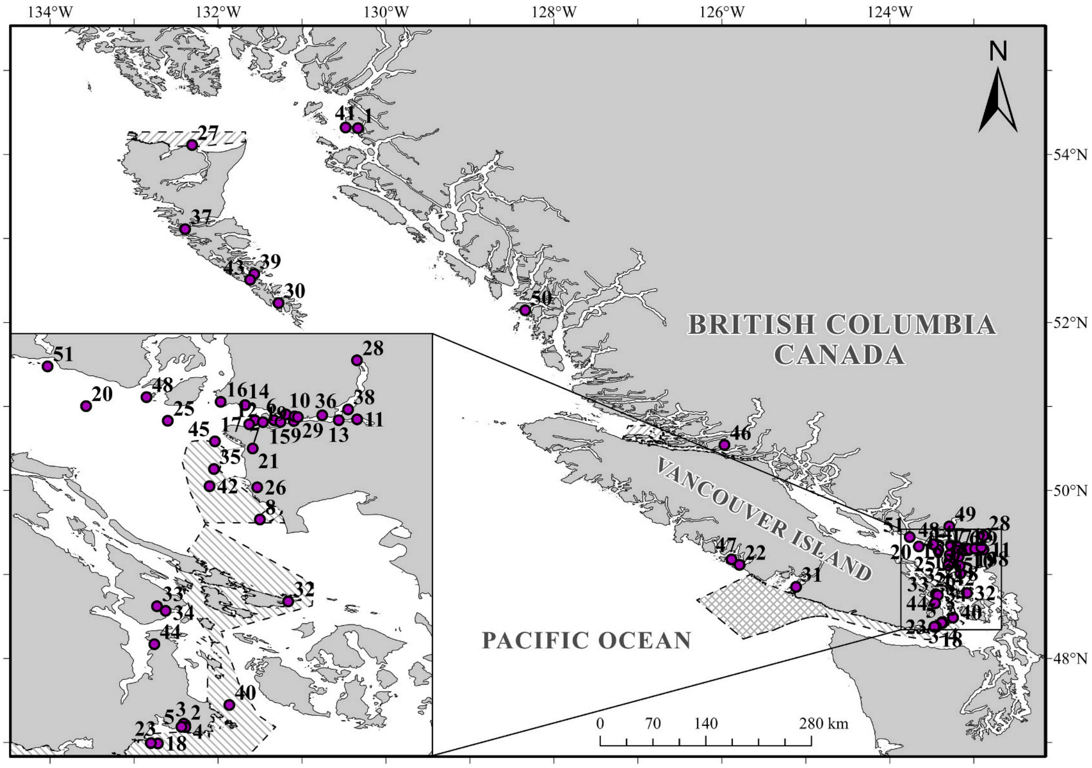

**Fig. 1.** Locations of 51 Pollution Tracker sampling sites along the coast of British Columbia, Canada, where subtidal surface sediment was collected. Sites are numbered with map IDs ordered from highest to lowest total polycyclic aromatic hydrocarbon concentrations. (Site names and corresponding map IDs are listed in Table A.1). Critical habitat of Northern Resident killer whales (///) and Southern Resident killer whales (\\\) are shown as hatched areas. The cross-hatched area represents shared critical habitat.

used to quantify 11 additional parent PAHs and 109 individual alkyl PAH isomers using the methodology described in Yunker et al. (2015). Example of chromatograms for reference samples, namely NIST 1597, a coal standard reference material from the Late Cretaceous Comox field, and Alberta Sweet Mixed Blend (ASMB) reference oil can also be found in Yunker et al. (2015).

### 2.4. Grain size and total organic carbon analyses

Total organic carbon (TOC) was determined by combustion using a carbon analyzer at ALS Environmental (B.C., Canada). Briefly, while total carbon (C) was determined by combustion and thermal conductivity detection, inorganic C was determined by weight loss after addition of hydrochloric acid. TOC was then calculated by the difference between these two measurements according to Nelson and Sommers (1996) and Loeppert and Suarez (1996).

Sediment particle size was determined at ALS Environmental (B.C., Canada). According to Burt (2009), dry sieving was used for coarse particles, wet sieving for sand particles and, finally, the pipette sedimentation method was used for clay particles.

### 2.5. Data analysis

Statistical analyses (calculation of standard deviation and averages) were carried out using R Statistical Software (Ver. 4.1.2; R Core Team, 2021).

A detailed evaluation of data quality was conducted by examining recoveries, blanks, detection limits, and resultant data profiles. Recoveries from the spiked sediment samples ranged from 39.1 to 101 % (average of 79 ± 14 %) with the majority (except the most volatile constituents) being within the quality assurance/quality control criteria of 70–130 % (Harris et al., 2011a). All data were recovery corrected. In the blanks, the majority of PAHs were below the detection limit with an average of only 16 PAHs detected out of the 75 measured. Levels detected in the blank never exceeded 2 nanograms per gram (ng/g) dry weight (dw). Blank values were subtracted from each sample value and the final data is presented in ng/g dw. To determine total PAH concentrations, analytes that were reported below the detection limit were considered to be zero.

Principal Components Analysis (PCA) was conducted to evaluate PAH patterns along the B.C. coast, quantifying the 75 PAHs as well as additional peaks. The stated PAH concentration was used for analytes reported by SGS AXYS Analytical as NDR (non-detectable range: peak detected but confirming-ion ratios outside specified range). Each PAH was evaluated for potential interferences, closeness to the limit of detection and the percentage of undetectable values before inclusion in the final PCA data set of 112 parent and alkyl PAHs.

The minor, unassigned alkyl PAH isomers for several C3 and C4 naphthalenes, methyl fluorenes, C3 phenanthrene/anthracenes, methyl fluoranthenes and methyl benz[*a*]anthracene/chrysenes that were assigned letters by Yunker et al. (2015) were uniformly undetectable in sediments from remote areas and these minor alkyl PAHs were all excluded from the PCA model. Undetectable values (455 or 8.0 % of the entire data set) were replaced by a random number between zero and the limit of detection by multiplying the value of the detection limit by a random number between 0 and 1.

Samples were normalised to the concentration total before PCA to remove artefacts related to concentration differences between samples. The centred log ratio transformation (division by geometric mean of the concentration-normalised sample followed by log transformation) was then applied to this compositional data set to produce a data set unaffected by negative bias or closure (Yunker et al., 2015) and where the average concentration and concentration total were identical for every sample. Data were then autoscaled before PCA to give every variable equal weight. Unsupervised PCA was performed using a program that employed the non-linear iterative partial least squares (NIPALS) algorithm using software provided by the Chemometrics Clinic (Seattle, Washington).

### 2.6. *Risk to the marine environment*

Two approaches were used to evaluate the risk posed by PAH levels in sediment along the BC coast.

First, PAH levels were compared to available sediment quality guidelines (SQGs) which provide scientific benchmarks for evaluating the potential for adverse biological effects in aquatic systems and are largely based on toxicity endpoints for invertebrates. In Canada, the Canadian Council of Ministers of the Environment (CCME) has developed marine SQGs including interim sediment quality guidelines (ISQGs) and probable effects levels (PELs) for 13 individual PAHs (CCME, 1999) (Table 1). The ISQG is the concentration below which adverse biological effects are expected to occur rarely while the PEL is the concentration above which adverse effects are expected to occur frequently (Environment Canada, 1998). In order to better assess the adverse effects related to the exposure to PAH mixtures, the mean probable effects level quotient (PEL-Q) was used (Long and MacDonald, 1998; Li et al., 2021). The PEL-Q was calculated as follows:

$$\text{Mean PEL} - \text{Q} = 1/n^{*}\Sigma\,(\text{C}_i/\text{PEL}_i),$$

where n is the number of measured PAHs with PEL available ($n = 13$), $C_i$ is the concentration of compound i in the sediment and $PEL_i$, the PEL of compound i in the sediment (Table A.1). Risk can then be classified as follows: mean PEL-Q $\leq 0.1$ indicates a potential risk, $>0.1$ but $<1$ indicates a moderate risk and $>1$ indicates a high risk (Li et al., 2021).

Second, sediment toxicity was calculated as toxic equivalents (TEQs) relative to BaP which represents the carcinogenic properties of a select number of PAHs ($n = 16$, Table A.1) through the activation of the aryl hydrocarbon receptor (AhR) (Villeneuve et al., 2002; Denison and Nagy, 2003). TEQs were calculated for each of the 16 EPA priority PAHs detected per site and summed together ($\sum$ TEQ). $\sum$ TEQ values were calculated as $\sum$ ($C_i$ x $TEF_i$), where $C_i$ = concentration compound i in sediment and $TEF_i$ = the toxic equivalency factor of compound i relative to BaP, as calculated in Li et al. (2021). TEF values were obtained from Nisbet and LaGoy (1992). Since $\sum$TEQ calculated for PAHs encompasses specific PAH compounds that have been shown to mediate their toxic effects through AhR, similar to dioxin-like compounds, Canadian SQGs for dioxin-like compounds (polychlorinated dibenzo-p-dioxins and polychlorinated dibenzofurans) have been deemed applicable for TEQs determined for PAHs (Pheiffer et al., 2018) and will be used in the present study (ISQG: 0.85 ng TEQ/kg; PEL: 21.5 ng TEQ/kg, (CCME, 2001).

## 3. Results and discussion

### 3.1. *Hydrocarbon concentrations in sediment*

PAHs were detected in sediment at all 51 sites, including sites in remote areas (Fig. 2). Total PAHs (75 analytes) ranged from 36 to 162,000 ng/g dw. The lowest PAH levels were detected in Sechelt (LG62 $_{\text{(map ID: 51)}}$) (36 ng/g dw), while samples from Prince Rupert Harbour (NWC1 $_{(1)}$) (162,000 ng/g dw) and four Victoria Harbour sites (VH1 $_{(3)}$, VH2 $_{(2)}$, VH3 $_{(4)}$, VH4 $_{(5)}$) (21,000–47,000 ng/g dw) had the highest PAH levels coast-wide. These observations are consistent with port activities, high vessel traffic, or historical operations in these harbours, such as the historical operation of a coal gas plant adjacent to Victoria Harbour (Yunker et al., 2012). Victoria Harbour alongside the East Terminal in Burrard Inlet are considered the most contaminated areas in the Canadian portion of the Salish Sea, based on a high-resolution analysis of additional contaminants of concern including legacy and emerging contaminants (Morales-Caselles et al., 2017b).

Our results are also consistent with hydrocarbon marine sediment studies in B.C. which assessed sediment contamination along a localized gradient of pristine to high vessel traffic/small spill areas. In those studies, the sum of parent and alkyl PAHs ranged from 11 to 27,050 ng/g dw (Harris et al., 2011a); 326–5560 ng/g (Yunker et al., 2014, 2015); and >260,000 ng/g dw (Yunker et al., 2011a), the latter being a contaminated site in Kitimat downstream of an active pulp and paper mill, and aluminum smelter.

**Table 1**
Percent (%) of the 51 sites that exceeded the marine interim sediment quality guidelines (ISQGs) and probable effect levels (PELs) established by the Canadian Council of Ministers of the Environment (CCME, 1999) for 13 individual PAHs (μg/kg dw) and for the sum of toxic equivalents ($\sum$ TEQs).

| PAHs of concern for environmental health | Risk or hazard statement | ISQG | % sites exceeding ISQG | PEL | % sites exceeding PEL |
|---|---|---|---|---|---|
| Low Molecular Weight PAHs: | | | | | |
| 2-Methylnaphthalene | Toxic to aquatic life with long lasting effects. | 20.2 | ***41.2*** | 201 | ***5.9*** |
| Naphthalene | Toxic to aquatic organisms. May cause long-term effects in the aquatic environment. Bioaccumulation of this chemical may occur along the food chain. | 34.6 | ***31.4*** | 391 | ***5.9*** |
| Acenaphthene | Very toxic to aquatic life with long lasting effects. | 6.71 | ***35.3*** | 88.9 | ***3.9*** |
| Acenaphthylene | Acute toxicity by inhalation, ingestion and dermal contact. | 5.87 | ***29.4*** | 128 | ***2.0*** |
| Anthracene | May cause cancer. Very toxic to aquatic life with long lasting effects. | 46.9 | ***29.4*** | 245 | ***9.9*** |
| Fluorene | Very toxic to aquatic organisms. May cause long-term adverse effects to aquatic life. | 21.2 | ***27.5*** | 144 | ***3.9*** |
| Phenanthrene | Very toxic to aquatic life with long lasting effects. | 86.7 | ***33.3*** | 544 | ***11.8*** |
| High molecular weight PAHs: | | | | | |
| Benz[*a*]anthracene | Might be mutagenic and carcinogenic. Very toxic to aquatic life with long lasting effects. | 74.8 | ***29.4*** | 693 | ***5.9*** |
| Benzo[*a*]pyrene | Mutagenic and carcinogenic. Very toxic to aquatic life with long lasting effects. May damage fertility. | 88.8 | ***27.5*** | 763 | ***5.9*** |
| Chrysene | Might be mutagenic and carcinogenic. Very toxic to aquatic life. May cause long lasting harmful effects to aquatic life. | 108 | ***29.4*** | 846 | ***7.8*** |
| Dibenz[*a,h*]anthracene | May be carcinogenic to aquatic organisms. Very toxic to aquatic life. Acute hazard with long lasting effects. | 6.22 | ***33.3*** | 135 | ***2.0*** |
| Fluoranthene | Toxic by all routes (i.e., inhalation, ingestion, dermal contact). Very toxic to aquatic life with long lasting effects. | 113 | ***31.4*** | 1494 | ***5.9*** |
| Pyrene | Very toxic to aquatic life with long lasting effects. | 153 | ***27.5*** | 1398 | ***5.9*** |
| $\sum$TEQ (ng TEQ/kg dw) | Variety of toxic effects through the activation of the aryl hydrocarbon receptor. | 0.85[a] | ***100*** | 21.5[a] | ***100*** |

[a] ISQG and PEL for $\sum$TEQs are those of polychlorinated dibenzo-p-dioxins/furans.

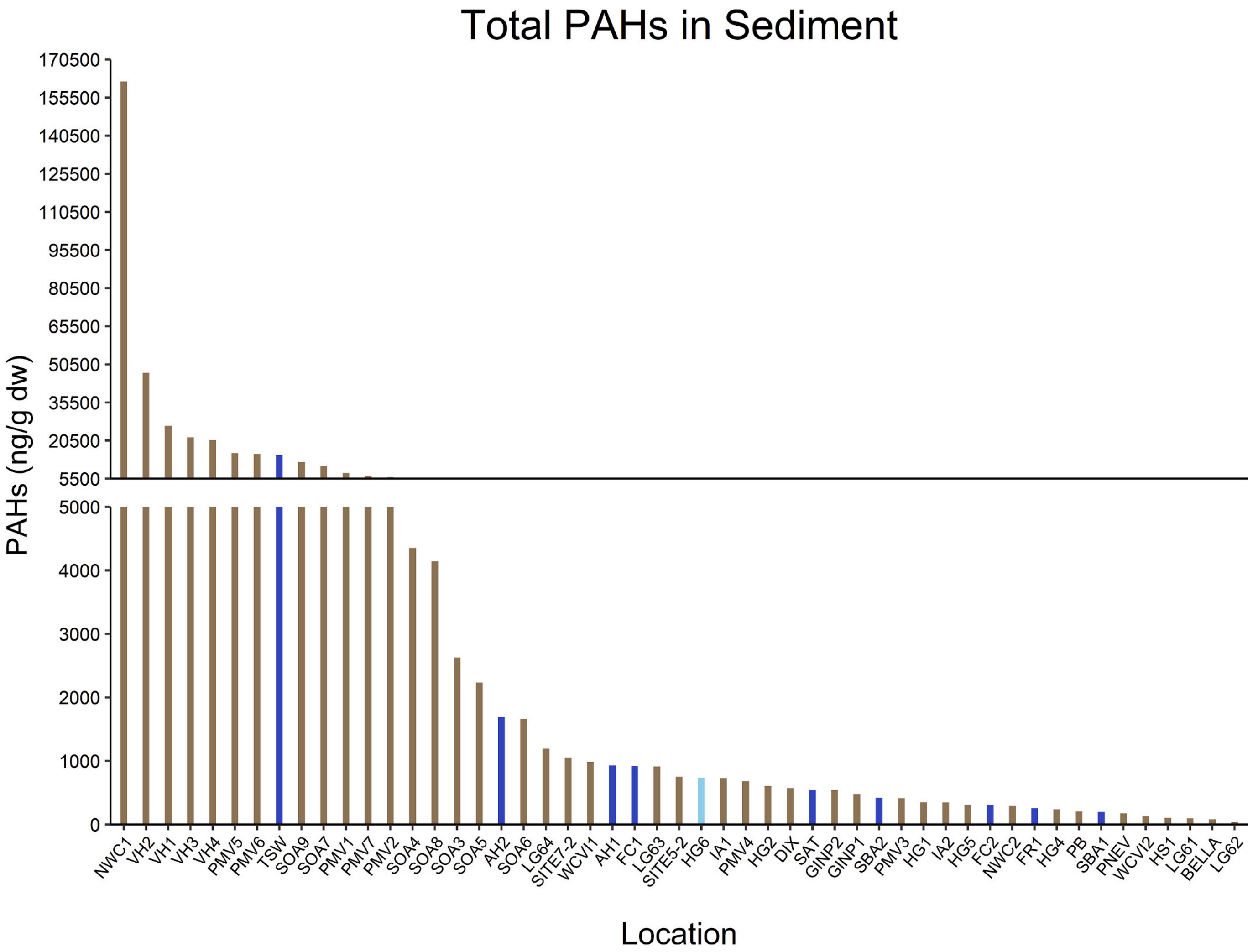

**Fig. 2.** Total polycyclic aromatic hydrocarbon (PAH) concentrations for subtidal sediment collected from 51 sites along the British Columbia coast between 2015 and 2017. Nine sites are within Southern Resident killer whale critical habitat (blue bars) while one site is within Northern Resident killer whale critical habitat (light blue bar). Sites are ordered by total PAH concentration from highest (left) to lowest (right). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Our analyses included the 16 priority PAHs used by the United States Environmental Protection Agency (EPA) (EPA, 2014) as they are often prioritized in monitoring and peer-reviewed literature. These 16 priority PAHs include naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benz[*a*]anthracene, chrysene, benzo[*b*]fluoranthene, benzo[*k*]fluoranthene, benzo[*a*]pyrene, benzo[*ghi*]perylene, indeno[1,2,3-*cd*]pyrene and dibenz[*a,h*] anthracene.

The sum of the 16 priority PAHs in our samples ranged from 10 ng/g dw (in Sechelt (LG62 $_{(51)}$)) to 67,000 ng/g dw (in Prince Rupert Harbour (NWC1 $_{(1)}$)). This concentration range was similar to recent levels reported in marine or coastal (estuaries, lagoons) sediments with 31–223 ng/g dw in the Siberian Arctic seas (Lakhmanov et al., 2022); 67–83 ng/g dw in the Persian Gulf (Zoveidadianpour et al., 2023); 5–76 ng/g dw in the Eastern Mediterranean (Darılmaz et al., 2013); 115–2398 ng/g dw in Tunisia (Mdaini et al., 2023); 12–389 ng/g dw in China (Li et al., 2023); and up to 60,300 ng/g dw in Vancouver, B.C., Canada following an oil spill (Morales-Caselles et al., 2017a). For levels in ports and harbours, up to 5361 ng/g dw were reported in the Flensburg Fjord at a German port (Wang et al., 2020), 336,770 ng/g dw in a Nova Scotia harbour (Davis et al., 2018), and up to 380,000 ng/g dw in Sydney Harbour, Australia (McCready et al., 2000).

Overall, the contribution of the 16 priority PAHs to total PAHs ranged from 5.4 % (in Armentieres Channel (HG1 $_{(37)}$)) to 47.7 % (in Grice Bay (WCVI1 $_{(22)}$)), contributing to, on average, 17 ± 3 % (SE) of the total PAHs detected (Fig. 3).

The bioavailability of PAHs in the aquatic environment is often associated with environmental variables such as particle size or total organic carbon (CCME, 1999). In the present study, total organic carbon (TOC) ranged from 0.2 to 12.3 % (Table A.1) and was not significantly correlated with total PAHs in sediment ($p = 0.352$). Significant and non-significant relationships have both been reported in the literature (Vane et al., 2020; Jesus et al., 2022; Yang et al., 2019) and the lack of consistency was attributed to the various type of carbon that can be present in the sediment with black carbon, coke, pitch and kerogen usually exhibiting higher PAH levels than predicted only based on the TOC content (Li et al., 2019; Jesus et al., 2022) A PCA was conducted to characterize sediment based on grain size composition (Fig. A.1). The first two principal components accounted for 92.8 % of the variance with PC1 and PC2 accounting for 65.3 % and 27.5 %, respectively. Sites associated with a high proportion of sand were positively associated with PC1 while sites associated with a higher content in silt and clay were negatively associated with PC1. There was minimal clustering along PC2 with only a few samples associating positively with PC2 and a higher proportion of gravel (PMV2; HS1; BELLA; NWC1 and NWC2). PC1 was negatively correlated with total PAHs in sediment ($R^2 = 0.11$; $p$

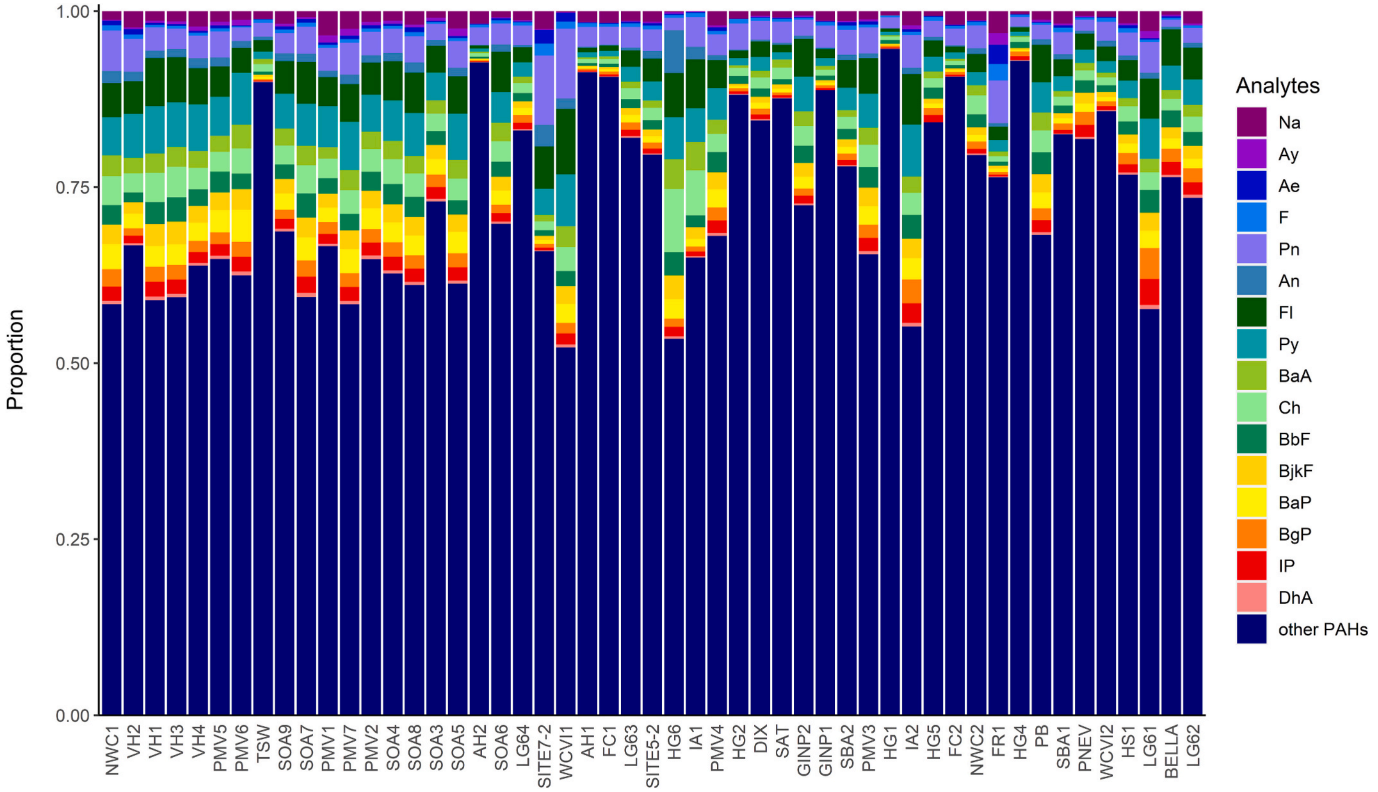

**Fig. 3.** The 16 EPA priority PAHs (naphthalene (Na), acenaphthylene (Ay), acenaphthene (Ae), fluorene (F), phenanthrene (Pn), anthracene (An), fluoranthene (Fl), pyrene (Py), benz[*a*]anthracene (BaA), chrysene (Ch), benzo[*b*]fluoranthene (BbF), benzo[*jk*]fluoranthenes (BjkF), benzo[*a*]pyrene (BaP), benzo[*ghi*]perylene (BgP), indeno[1,2,3-*cd*]pyrene (IP), and dibenz[*a*,*h*]anthracene (DhA)), contributed to an average of 17 ± 3 % (SE) of the total PAHs detected. Sites are ordered by total PAH concentration from highest (left) to lowest (right).

$< 0.05$) indicating that sites heavy in sand had the highest PAH concentrations similar to what has been reported previously (Gnandi et al., 2011).

### 3.2. PAH patterns along the B.C. coast

The bioavailability of PAHs can vary substantially between solid fuel and its combustion products (coal, coke, wood char), the products of liquid fossil fuel combustion, and liquid petroleum products. It is therefore necessary to establish the major combustion and petrogenic sources – and if possible their relative contribution to samples from different areas – to be able to estimate the PAH bioavailability (Yunker et al., 2015, 2011a; Short et al., 1999). To this end, PAH patterns along the coast were evaluated using a PCA model.

The first two principal components accounted for 42.3 % and 11.5 % of variance, respectively, for PC1 and PC2, so that the model shown in Fig. 4 accounted for 53.8 % of the variance in the data set. In the PCA loadings plot (Fig. 4a), the parent and alkyl PAH variables generally clustered by compound type, where the parent PAHs and the alkyl fluoranthene/pyrenes and alkyl benz[*a*]anthracene/chrysenes project on the left side, primarily from the lower centre to the upper left, and the alkyl naphthalenes and alkyl phenanthrene/anthracenes primarily project from the centre right to the upper right side. For the two variables that were only detectable in urban samples, indeno[1,2,3-*cd*]fluoranthene projects close to the other 276 Da parent PAHs such as benzo[*ghi*]perylene, while 1,2-dimethylphenanthrene projects well outside the main cluster of alkyl phenanthrene/anthracenes. The dibenzothiophenes and alkyl fluorenes, plus small numbers of parent PAHs and alkyl PAH isomers, scattered across the bottom of the loadings plot with little apparent pattern. Here, the exception is the alkyl fluorenes, which all project on the lower right.

Samples from urban and industrial areas including Victoria Harbour (VH1$_{(3)}$, VH2$_{(2)}$, VH3$_{(4)}$, VH4$_{(5)}$), Burrard Inlet (Vancouver Harbour; PMV1$_{(11)}$, PMV2$_{(13)}$, PMV3$_{(36)}$, PMV4$_{(29)}$, PMV5$_{(6)}$, PMV6$_{(7)}$, PMV7$_{(12)}$, SOA4$_{(14)}$, SOA5$_{(17)}$, SOA6$_{(19)}$, SOA7$_{(10)}$, SOA8$_{(15)}$, SOA9$_{(9)}$), Indian Arm (IA1$_{(28)}$, IA2$_{(38)}$), and Howe Sound (LG61$_{(49)}$, HS1$_{(48)}$, SOA3$_{(16)}$) all projected on the left side of the PC1, while samples from the Fraser River (FR1$_{(42)}$, SBA1$_{(45)}$, SBA2$_{(35)}$, SITE5-2$_{(26)}$, SITE7-2$_{(21)}$), southern Vancouver Island (GINP1$_{(34)}$, GINP2$_{(33)}$, AH1$_{(23)}$, AH2$_{(18)}$, FC1$_{(24)}$, FC2$_{(40)}$, DIX$_{(31)}$, WCVI2$_{(47)}$) and the Strait of Georgia (LG62$_{(51)}$, LG63$_{(25)}$, LG64$_{(20)}$, TSW$_{(8)}$, SAT$_{(32)}$) almost all projected on the right side (Fig. 4b). Four of the five samples collected around Haida Gwaii projected on the lower right side of PC1 (HG1$_{(37)}$, HG2$_{(30)}$, HG4$_{(43)}$, HG5$_{(39)}$). Where samples from an area projected on both sides of PC1, the samples on the left side were from harbour or more urbanised areas, namely Patricia Bay in Saanich Inlet north of Victoria (PB$_{(44)}$), Grice Bay from the West Coast (WCVI1$_{(22)}$), Wiah Point from Haida Gwaii (HG6$_{(27)}$), Bella Bella (BELLA$_{(50)}$), and Prince Rupert Harbour from the North Coast (NWC1$_{(1)}$).

The PCA model showed that, generally, urban locations had the highest proportion of parent PAHs, alkyl fluoranthene/pyrenes and alkyl benz[*a*]anthracene/chrysenes. Indeed, C1-fluoranthenes/pyrenes were dominant at twenty-one urban sites including: Victoria Harbour (VH1$_{(3)}$, VH2$_{(2)}$, VH3$_{(4)}$, VH4$_{(5)}$), Patricia Bay (PB$_{(44)}$), Indian Arm (IA2$_{(38)}$), Wiah Point (HG6$_{(27)}$), Prince Rupert Harbour (NWC1$_{(1)}$) and Burrard Inlet (PMV1$_{(11)}$, PMV2$_{(13)}$, PMV3$_{(36)}$, PMV4$_{(29)}$, PMV5$_{(6)}$, PMV6$_{(7)}$, PMV7$_{(12)}$, SOA4$_{(14)}$, SOA5$_{(17)}$, SOA6$_{(19)}$, SOA7$_{(10)}$, SOA8$_{(15)}$, SOA9$_{(9)}$). Additionally, PCA results showed that, in general, remote and less-urbanised locations contained higher proportions of alkyl naphthalenes, phenanthrene/anthracenes and fluorenes.

Similar PAH composition patterns and PCA results were observed in grab samples from B.C. near-shore ocean sediment collected in 2006 (nine samples from Eastern Vancouver Island with a depth of 126–380 m) and 2007 (eight samples from Vancouver Harbour with a depth of

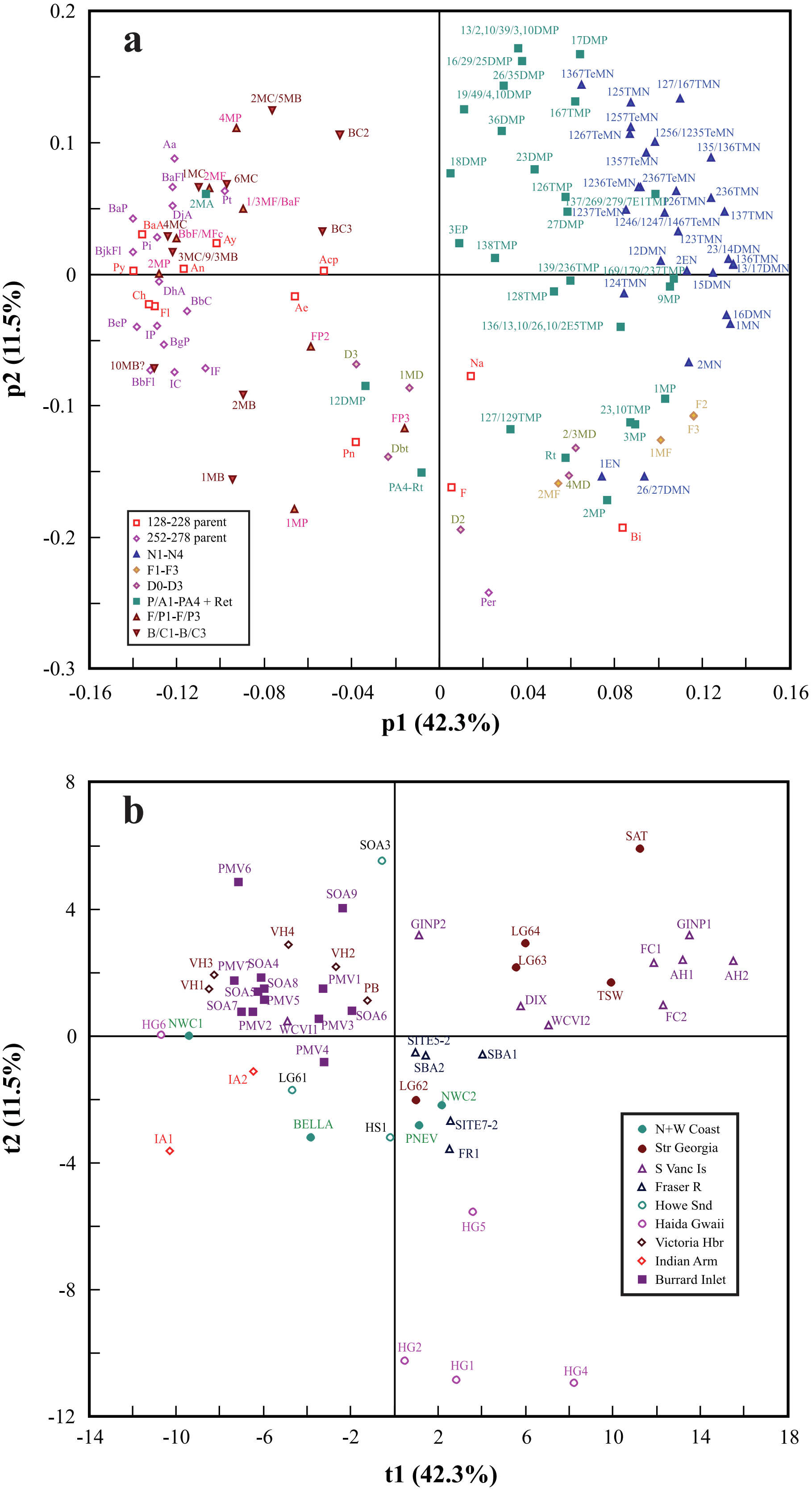

**Fig. 4.** a) Variable projections for p1 vs. p2 in the PCA loadings plot for a PCA model using 112 variables (parent and alkyl PAHs; see Table A.2 for variables and abbreviations). Variables are separated by compound type for clarity into the following groups: the two to four ring parent PAHs (128–228 Da), five and six ring parent PAHs (252–278 Da), alkyl naphthalenes (N1–N4), alkyl fluorenes (F1–F3), dibenzothiophenes (D0–D3), alkyl phenanthrene/anthracenes (P/A1–P/A4) and retene (Rt), alkyl fluoranthene/pyrenes (F/P1–F/P3), and alkyl benz[*a*]anthracene/chrysenes (B/C1–B/C3). b) Sample projections for t1 vs. t2 in the PCA scores plot for the 51 sediment grab samples.

1–35 m) (Yunker et al., 2015; Harris et al., 2011a). This may indicate minimal variation over time in PAH composition in surface sediment collected from these areas of coastal B.C.

### 3.3. Parent PAH composition as an indicator of combustion inputs

The principal four, five and six ring PAHs with molecular masses of 202 (fluoranthene, pyrene), 252 (benzo[*b/j/k*]fluoranthene, benzo[*e*]pyrene), and 276 (indeno[1,2,3-*cd*]pyrene, benzo[*ghi*]perylene) Da primarily have a combustion source in B.C. aquatic environments (Yunker et al., 2002, 2015). These six parent PAHs can be arranged into three specific ratios which can distinguish between petroleum, and the combustion of liquid fuel (vehicle fuel and crude oil), and biomass/solid fuel (coal, wood, or grass; Yunker et al., 2002, 2011b, 2015).

Petroleum, liquid fuel combustion, and biomass/solid fuel combustion source boundaries are defined in Yunker et al. (2002, 2015). For the ratio of fluoranthene to fluoranthene plus pyrene (Fl/(Fl + Py)), a calculated ratio value below the petroleum boundary of 0.4 indicates a petroleum source, while values between 0.4 and 0.5 indicate liquid fuel combustion sources, and values greater than 0.5 indicate biomass/solid fuel combustion sources (Yunker et al., 2002). For the ratio of benzo[*b/j/k*]fluoranthene to benzo[*b/j/k*]fluoranthene plus benzo[*e*]pyrene (BFl/(BFl + BeP)), ratios below the petroleum boundary of 0.5 (Fig. 5) indicate a petroleum source, while values between 0.5 and 0.7 indicate liquid fuel combustion, and those over 0.7 indicate biomass/solid fuel combustion. For the ratio of indeno[1,2,3-*cd*]pyrene to indeno[1,2,3-*cd*]pyrene plus benzo[*ghi*]perylene (IP/(IP+(BgP)), ratios below the petroleum boundary of 0.2 (Fig. 5) indicate a petroleum source, while values between 0.2 and 0.5 indicate liquid fuel combustion, and those over 0.5 indicate biomass/solid fuel combustion.

For each of the 51 Pollution Tracker sediment samples, all of the BFl/(BFl + BeP) and IP/(IP + BgP) ratios, and all but one of the Fl/(Fl + Py) ratios, were above the petroleum boundaries (Fig. 5a,b), indicating a dominance of combustion-related sources. The sole exception for Fl/(Fl + Py) was Burrard Inlet sample PMV6 $_{(7)}$ (Fig. 5a), which had a ratio of 0.32, indicating a petroleum source. Ratios for ASMB (Alberta Sweet Mixed Blend) oil and Comox coal fell well outside the sediment sample range, and inside the range typical for fossil fuels (lower left of Fig. 5a and b).

Previous analysis of sediment in coastal B.C. also demonstrated that combustion products are the predominant source of four ring and higher parent PAHs with molecular weights of 202, 252 and 276 Da, with most samples also close to the borderline between biomass/solid fuel combustion and liquid fuel combustion based on calculated values for the same three ratios used for our samples (Yunker et al., 2015). Additionally, analysis of Vancouver Harbour sediment using the two ratios Fl/(Fl + Py) and IP/(IP + BgP) also demonstrated samples were on the border between both biomass/solid fuel combustion and liquid fuel combustion (Yunker et al., 2002). Similar results were found in Nova Scotia harbours, where these same three diagnostic ratios indicated both petroleum combustion and biomass/solid fuel combustion sources in sediments (Davis et al., 2019). Our results are consistent with the top contributors to PAH emissions in Canada (Berthiaume et al., 2021; Marvin et al., 2021) which are from biomass/solid fuel combustion and liquid fossil fuel combustion.

### 3.4. Petrogenic source apportionment using alkyl naphthalene isomers

The proportion of alkyl PAHs found in oil and coal is greater than the proportion of parent PAHs (Yunker et al., 2015). In particular, the C2 – C4 naphthalenes make up a large proportion of the alkyl PAH profile found in both ASMB oil and coal from the Late Cretaceous Comox field (Yunker et al., 2015). The C3 and C4 naphthalenes retain much of their original petrogenic source signature of coal and oil inputs to sediments because, when in oil, they are resistant to biodegradation (Farias et al., 2008; Greenwood et al., 2008; Chen et al., 2013), and, when in coals, they are protected by the solid matrix (Short et al., 1999; Cornelissen et al., 2005; Jonker et al., 2005; Yunker et al., 2011a). The largest differences in isomer composition between oil and coal are shown by the isomers 1,2,5-TMN (trimethylnaphthalene) and 1,2,5,6/1,2,3,5-TeMN (tetramethylnaphthalene; Yunker et al., 2015). Ratios of 1,2,5-TMN and 1,2,5,6/1,2,3,5-TeMN compared with the major isomer in each series (1,3,7-TMN and 1,3,6,7-TeMN, respectively) show the largest differences between the samples, and are shown in (Fig. 6).

In Fig. 6, ASMB oil plotted in the lower left and the two samples of Comox coal plotted on the right. Accordingly, the three samples plotting on the lower left including PMV3 $_{(36)}$ and SOA6 $_{(19)}$ from Burrard Inlet and Tsawwassen (TSW $_{(8)}$) exhibit the strongest influence of petroleum products in accordance with their location near Canada's largest port, which receives 3000 ships annually (Port of Vancouver, 2025). In particular, site TSW $_{(8)}$ is a container ship port, and is expected to expand its container capacity in the future (Port of Vancouver, 2024). Additionally, sample IA1 $_{(28)}$ from Indian Arm on the lower centre had a low TeMN ratio with a higher TMN ratio, and does not fit within the trend line for other samples, but petrogenic input is still suggested.

Samples plotting on the upper left in Fig. 6 have an oil influence that is more pronounced for the trimethylnaphthalenes than for the tetramethylnaphthalenes. These include the Haida Gwaii samples from Armentieres Channel and Louscoone Inlet (HG1 $_{(37)}$ and HG2 $_{(30)}$), Bella Bella (BELLA $_{(50)}$), Metlakatla (NWC2 $_{(41)}$), and two Burrard Inlet samples (PMV2 $_{(13)}$ and SOA7 $_{(10)}$).

Samples plotting on the upper right of Fig. 6 have the strongest coal influence. The two with the highest coal ratios are Wiah Point (HG6 $_{(27)}$) on northern Graham Island in Haida Gwaii, and Howe Sound 3 (SOA3 $_{(16)}$) near West Vancouver. Other samples with high alkyl naphthalene ratios include Burrard Inlet 14 (SOA9 $_{(9)}$), Burrard Inlet 6 (PMV6 $_{(7)}$) (which also had a low Fl/(Fl + Py) ratio), Sechelt (LG62 $_{(51)}$), Patricia Bay (PB $_{(44)}$), and Victoria Harbour 2 (VH2 $_{(2)}$). Cenozoic lignites (young coal deposits) from the Skonun Formation, which underlies the eastern third of Graham Island, outcrop along the coast near to where HG6 $_{(27)}$ was collected (Sutherland Brown, 1968; Yunker et al., 2014), explaining the strong coal influence in that sample.

The other samples with high coal ratios are likely to be either locations of former coal loading facilities or have experienced coal spills or coal dumping in the past, such as the historical operation of the Rock Bay coal gas plant (now a remediated site) that operated from 1862 through 1952 in upper Victoria Harbour (Yunker et al., 2012), closest to sites Victoria Harbour 1 (VH1 $_{(3)}$) and Victoria Harbour 2 (VH2 $_{(2)}$). Because the PAHs associated with coal are more stable, expected to be more persistent in the marine environment, and accumulate in sediment (Yunker et al., 2015, 2002, 2012), they are expected to be less bioavailable to marine fauna than the PAHs associated with liquid fuel combustion. However, most samples in Fig. 6 have values between the oil and coal references, indicating mixed sources of both.

### 3.5. Risk to the marine environment

With contaminants being one of the main threats to SRKW and their main prey, Chinook salmon (*Oncorhyncus tshawytscha*) (ECCC, 2020), understanding marine environmental quality is key. Stomach contents of juvenile Chinook salmon include a variety of benthic invertebrate species (Duguid et al., 2021; Kennedy et al., 2018), and these food contents have been found to contain elevated concentrations of PAHs, up to ten-fold higher than PAH concentrations in nearby sediment (Yanagida et al., 2012). This may reflect the inability of benthic invertebrates to metabolize hydrocarbons sufficiently, which varies by species (Meador, 2003). Therefore, we can expect that SRKW feeding on salmon in their critical habitat would experience sporadic and short-term exposure to PAHs (Grant and Ross, 2002) through the ingestion of contaminated prey (Ball and Truskewycz, 2013).

Since marine mammals can metabolize PAHs through the mixed function oxidase system as reviewed in Ruberg et al. (2021), the

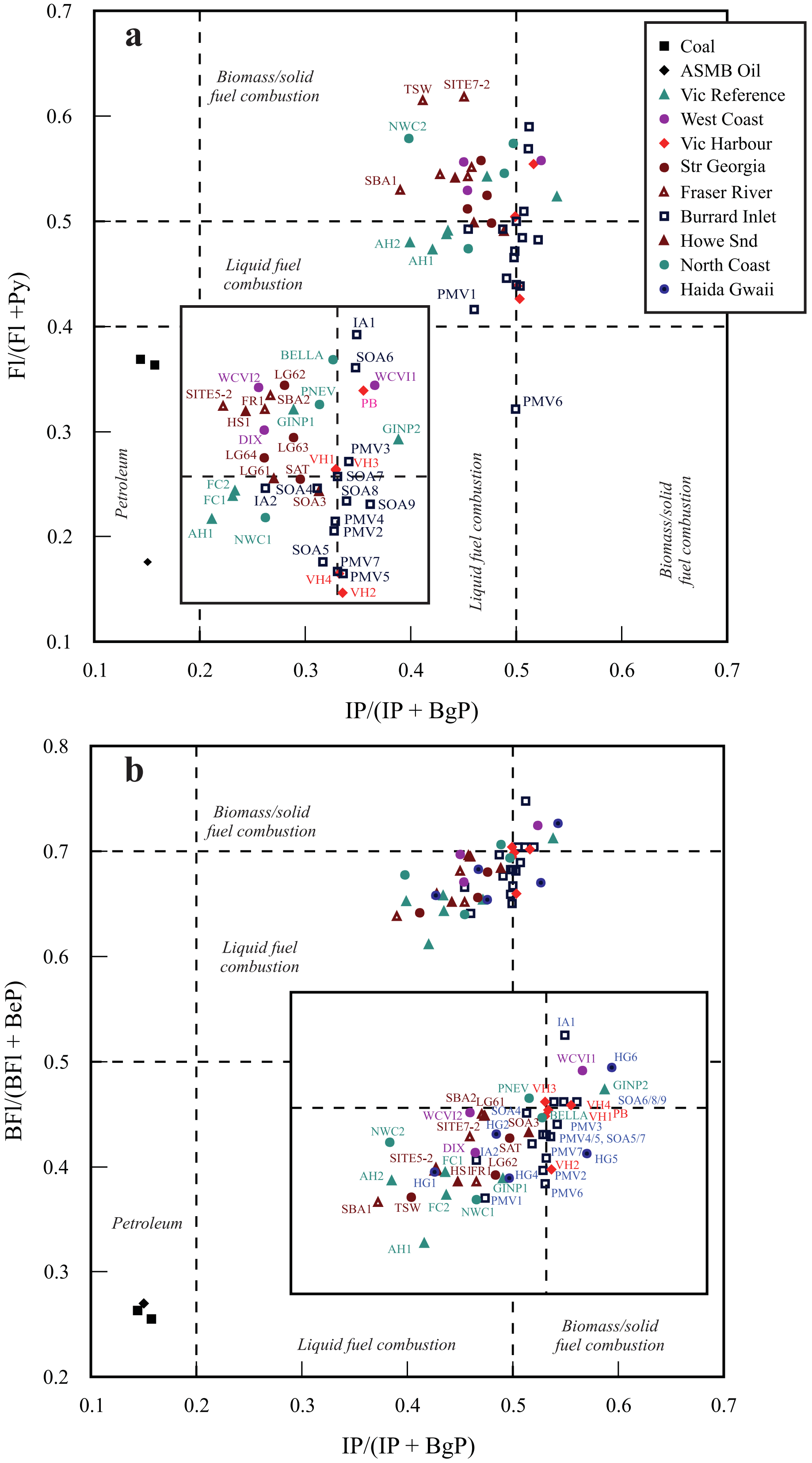

**Fig. 5.** PAH ratio cross plots showing proportions of a) fluoranthene to fluoranthene plus pyrene Fl/(Fl + Py) and b) benzo[b/j/k]fluoranthene to benzo[*e*]pyrene (BFl/(BFl + BeP)) vs. indeno[1,2,3-*cd*]pyrene to benzo[*ghi*]perylene (IP/(IP + BgP)). Source boundaries are from Yunker et al., 2002, 2011b, 2015.

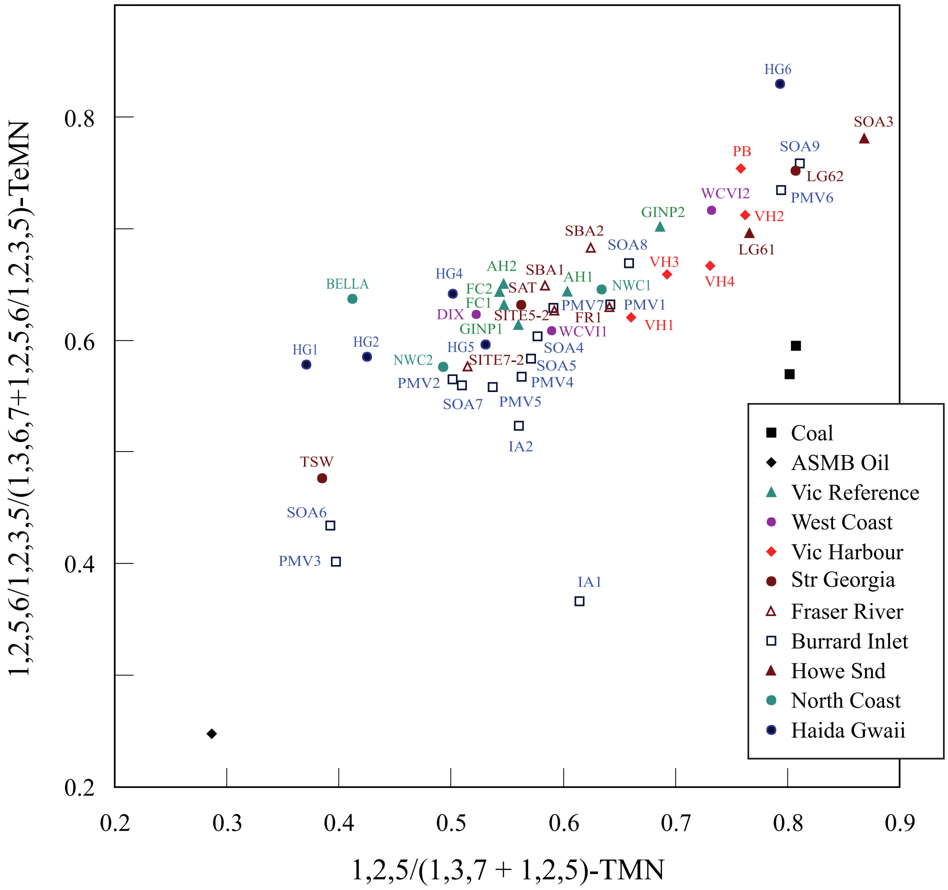

**Fig. 6.** C3 and C4 naphthalene cross plots show ratios of 1,2,5-TMN (trimethylnaphthalene) and 1,2,5,6/1,2,3,5-TeMN (tetramethylnaphthalene), compared with the major isomer in each series (1,3,7-TMN, and 1,3,6,7- TeMN, respectively).

detection of low levels of PAHs in marine mammal tissues may reflect recent exposure to PAHs (Lee et al., 2023). However, the health consequences of such sporadic and short-term exposures to PAHs are unclear and obfuscated by SRKW exposure to many other contaminants of notable concern, including very high levels of biomagnifying polychlorinated biphenyls, polybrominated diphenyl ethers, and legacy pesticides.

PAH levels were compared to SQGs developed by the CCME for 13 individual PAHs. While these guidelines do not take mixtures of PAHs into account, and only provide guidelines for the protection of invertebrates, they can provide some insight into the quality of the marine environment. The low molecular weight PAHs listed in Table 1 are considered acutely toxic but noncarcinogenic to aquatic organisms, and field studies with invertebrates suggest that for these low molecular weight PAHs mortality and sublethal effects occur at levels higher than the ISQGs (CCME, 1999). Conversely, the high molecular weight PAHs are considered not acutely toxic to aquatic organisms, but some are carcinogenic. Field studies with invertebrates and fish suggest that for these high molecular weight PAHs, mortality and sublethal effect occur at levels higher than the PELs (CCME, 1999).

Of the 51 Pollution Tracker sites, half (51 %) exceeded the CCME marine ISQGs for at least one of the 13 PAHs, while 14 % exceeded CCME marine PELs for at least one of the 13 PAHs. Additionally, 25 % of the sites exceeded ISQGs for all 13 PAH analytes, including sites from Victoria Harbour ((VH1 $_{(3)}$, VH2 $_{(2)}$, VH3 $_{(4)}$, VH4 $_{(5)}$)), Burrard Inlet ((PMV1 $_{(11)}$, PMV2 $_{(13)}$, PMV5 $_{(6)}$, PMV6 $_{(7)}$, PMV7 $_{(12)}$, SOA7 $_{(10)}$, SOA8 $_{(15)}$, SOA9 $_{(9)}$)), and Prince Rupert Harbour (NWC1 $_{(1)}$). Of these, sites VH1 $_{(3)}$ and VH2 $_{(2)}$ exceeded PELs for seven and eleven of the 13 PAHs, respectively, while site NWC1 $_{(1)}$ exceeded PELs for all 13 PAHs.

Of the 13 PAH analytes with available guidelines, benzo[*a*]pyrene (BaP) is considered the most toxic compound. Concentrations of BaP coast-wide ranged between 1 and 5040 ng/g dw (Fig. 3) with the highest concentration detected at Prince Rupert Harbour (NWC1 $_{(1)}$). 27.5 % of Pollution Tracker sites exceeded the ISQG for BaP (88.8 ng/g dw) while 5.9 % exceeded the PEL for BaP (763 ng/g dw) (Table 1). Similarly, Harris et al. (2011a) reported 42 % of their B.C. sediment samples exceeded the ISQG for BaP, while 5 % exceeded the PEL.

When calculating the mean PEL-Q values (Table A.1) to evaluate the combined biological effects of PAHs in sediment along the B.C. coast, we found that mean PEL-Q ranged from 0.001 at Sechelt (LG62 $_{(51)}$) to 7.6 at Prince Rupert Harbour (NWC1 $_{(1)}$). 69 % of the sites had values <0.1 suggesting only a potential risk associated with PAH exposure from sediment, 25 % of the sites had mean PEL-Q values indicating moderate risk, and two sites were categorized as having a high risk of biological effects with values >1 (Prince Rupert Harbour (NWC1 $_{(1)}$), and Victoria Harbour 2 (VH2 $_{(2)}$)).

$\sum$ TEQs (Table A.1) ranged from 8404 ng TEQ/g dw at Prince Rupert Harbour (NWC1 $_{(1)}$) to 0.94 ng TEQ/g dw at Armentieres Channel (HG1 $_{(37)}$) and Sechelt (LG62 $_{(51)}$). $\sum$ TEQs were higher in sediments collected from harbours, bays and inlets associated with urban or industrial activities than in samples from more remote and less-urbanised sites (Wilcoxon Test, $p < 0.05$), indicating the higher carcinogenic potency of sediments from urban and industrial areas. When compared to the CCME SQGs for dioxin-like compounds, $\sum$ TEQs for all 51 Pollution Tracker sites exceeded both the ISQG of 0.85 ng TEQ/kg and the PEL of 21.5 ng TEQ/kg (Table 1). The TEQ values reported here were extremely high compared to the guidelines, similar to what was reported in Pheiffer et al. (2018) and they highlight the fact that the CCME includes a safety factor of 10 in their guideline derivation to increase their protective function. With the safety factor removed, 43 % of the samples were below the ISQG and 76 % were below the PEL.

Of the 51 Pollution Tracker sites, nine are in SRKW critical habitat (includes the transboundary waters of the southern Strait of Georgia, Haro Strait, Juan de Fuca Strait, and Southwestern Vancouver Island (Fisheries and Oceans Canada (DFO), 2018)), one is in NRKW critical habitat (includes the waters of Johnstone Strait and southeastern Queen Charlotte Strait, (DFO, 2018)), and all are within NRKW and SRKW range (Fig. 2).

Within the nine SRKW critical habitat sites, four sites exceeded the ISQG for 2-methylnaphthalene including Tsawwassen (TSW $_{(8)}$), Albert Head 1 (AH1 $_{(23)}$), Albert Head 2 (AH2 $_{(18)}$), and Finnerty Cove 1 (FC1 $_{(24)}$). Additionally, Fraser River 1 (FR1 $_{(42)}$) exceeded the ISQG for acenaphthene. Furthermore, Tsawwassen (TSW $_{(8)}$) exceeded ISQGs for eight additional PAHs including anthracene, benz[*a*]anthracene, chrysene, dibenz[*a,h*]anthracene, fluoranthene, naphthalene, phenanthrene, and pyrene, and, exceeded the PEL for 2-methylnaphthalene. Levels of hydrocarbons at the one NRKW critical habitat site did not exceed marine CCME ISQGs. Percent of the 13 individual PAHs with available CCME sediment quality guidelines within the overall total PAHs detected at sites within NRKW and SRKW critical habitat are shown in Fig. 7.

Of the ten PAHs that exceeded sediment quality guidelines at Pollution Tracker sites within SRKW critical habitat, nine including 2-methylnaphthalene, acenaphthene, anthracene, benz[*a*]anthracene, chrysene, fluoranthene, naphthalene, phenanthrene, and pyrene were detected previously in a stranded SRKW mother and had transferred in-utero to the fetus (Lee et al., 2023).

The earlier ratio analyses suggested a mixture of biomass/solid fuel combustion sources and liquid fuel combustion sources (Fig. 5a,b), as well as petroleum sources (Fig. 6) for PAHs detected in sediment at Tsawwassen (TSW $_{(8)}$). Since the PAHs in both petroleum and liquid fossil fuel combustion are predicted to be fully bioavailable to marine fauna (Yunker et al., 2015), this is of concern for SRKW. Diagnostic ratios of PAHs detected in liver and skeletal muscle samples collected from stranded SRKWs that were found on the coast of Vancouver Island and in the Strait of Georgia, indicated PAH sources of liquid fossil fuel combustion and petroleum combustion (Lee et al., 2023). While Tsawwassen (TSW $_{(8)}$) is within SRKW critical habitat, it is also the site of a major marine transportation centre, recently approved for expansion. With the expansion which will increase the port's container terminal capacity by nearly 50 % (Port of Vancouver, 2023), vessel traffic is expected to increase, along with vessel associated petroleum discharges and liquid fuel combustion.

At the remaining sites with guideline exceedances in SRKW critical habitat, Fig. 6 shows the major sources of PAHs are from oil (Burrard Inlet 3 (PMV3 $_{(36)}$), Burrard Inlet 11 (SOA6 $_{(19)}$)), or both coal and oil (Fraser River 1 (FR1 $_{(19)}$), Albert Head 1 (AH1 $_{(23)}$), Albert Head 2 (AH2 $_{(18)}$), and Finnerty Cove 1 (FC1 $_{(24)}$)), while Fig. 5a,b shows these sites also contain mixtures of both biomass/solid fuel combustion sources, and liquid fuel combustion sources.

## 4. Conclusion

Our dataset strengthens the understanding of the PAH distribution in the coastal B.C. environment. We add to existing data on PAHs in Burrard Inlet and the Fraser River, and provide new information on PAHs and their sources in Howe Sound, Victoria Harbour, the West coast of Vancouver Island, the North Coast, and Haida Gwaii.

While natural petrogenic sources do contribute to the PAH profile in samples from Hecate Strait and Queen Charlotte Sound, we found that combustion is the primary contributor to the PAH sediment profile along the B.C. coast, with most Pollution Tracker samples showing mixed

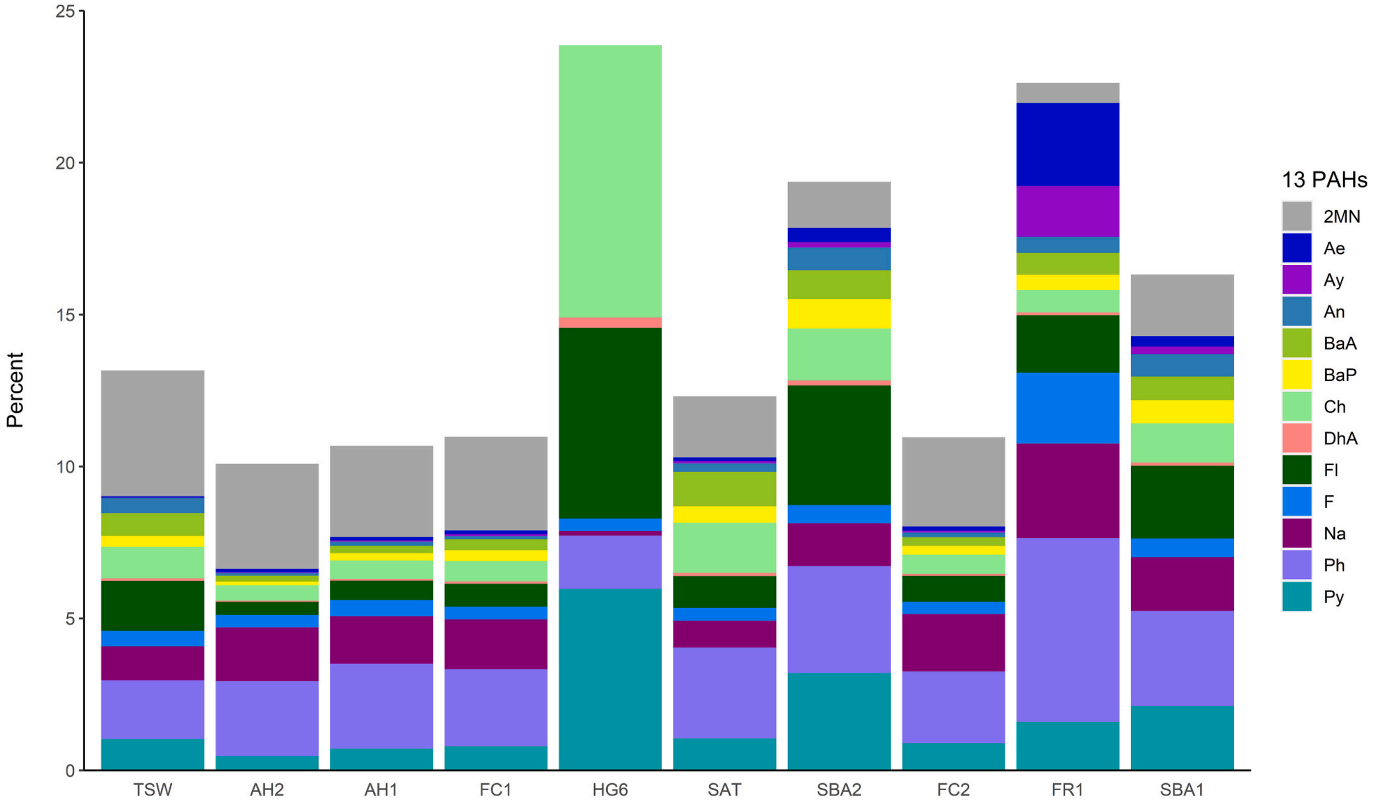

**Fig. 7.** Percent of the 13 individual PAHs with available CCME sediment quality guidelines: 2-methylnaphthalene (2MN), acenaphthene (Ae), acenaphthylene (Ay), anthracene (An), benz[*a*]anthracene (BaA), benzo[*a*]pyrene (BaP), chrysene (Ch), dibenz[*a,h*]anthracene (DhA), fluoranthene (Fl), fluorene (F), naphthalene (Na), phenanthrene (Ph), and pyrene (Py) within the overall total PAHs detected in sediment at nine sites within Southern Resident killer whale (SRKW) critical habitat and one site within Northern Resident killer whale (NRKW) critical habitat. Sites are ordered by overall total PAH concentration from highest (left) to lowest (right).

sources of both biomass/solid fuel combustion and liquid fuel combustion. Both sources of combustion are expected to increase in the future due to increased shipping, urban development and population growth in coastal areas, as well as the capacity for climate change to increase the severity and frequency of forest fires (Muir and Galarneau, 2021).

Of the sites within SRKW critical habitat, eight were categorized as having potential ecological risk associated with PAH exposure from sediment, while site Tsawwassen (TSW $_{(8)}$) was categorized as having moderate ecological risk. The detected mixtures of biomass/solid fuel combustion sources, liquid fuel combustion sources, and coal and oil sources in SRKW critical habitat indicate complex inputs of PAHs to these sediments. In particular, those PAHs from liquid fuel combustion and oil pose a risk to both SRKW and their prey, because they are expected to be fully bioavailable to the marine food web.

With increased industrialization on B.C.'s coast, such as the recently completed Trans Mountain Expansion project set to double the volume of oil that is shipped through SRKW range and critical habitat, risk of a large catastrophic oil spill is higher than ever. Our high-resolution baseline hydrocarbon data is also a valuable tool for post-spill event assessment.

## CRediT authorship contribution statement

**Elizabeth J. Ruberg:** Writing – original draft, Visualization, Formal analysis. **Mark B. Yunker:** Writing – review & editing, Visualization, Methodology, Formal analysis. **Gabriela Aguirre-Martinez:** Writing – review & editing. **Kelsey Delisle:** Writing – review & editing. **Carmen Morales-Caselles:** Writing – review & editing, Funding acquisition, Conceptualization. **Peter S. Ross:** Writing – review & editing, Funding acquisition, Conceptualization. **Marie Noël:** Writing – review & editing, Visualization, Funding acquisition, Conceptualization.

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## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.marpolbul.2025.118180.

## Data availability

Data will be made available on request.

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