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# **Special Collection:**

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#### **Key Points:**

- Sedimentary polycyclic aromatic hydrocarbons (PAHs) in the East Siberian Arctic Shelf (ESAS) have a mixture of petrogenic and pyrogenic sources with distinct input pathways
- Permafrost erosion serves as a significant input pathway and PAHs contributor in the rapidly changing Arctic coasts
- Black carbon plays a key role in the behavior and sequestration of pyrogenic PAHs in the Arctic coastal margins

#### **Supporting Information:**

Supporting Information may be found in the online version of this article.

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# Sources and Fates of Sedimentary Polycyclic Aromatic Hydrocarbons in the East Siberian Arctic Shelf: Implications for Input Pathways and Black Carbon Constraint

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**Abstract** The Arctic region is experiencing more rapid climate changes than the other parts of the world and serves as a sink for semi-volatile persistent organic pollutants, such as polycyclic aromatic hydrocarbons (PAHs), which can be utilized as molecular markers for pyrogenic carbon, such as black carbon (BC). As the sea ice retreats and increased terrestrial inputs with widespread wildfires, the PAH concentrations in the Arctic Ocean are rising. In this study, the sources and fates of PAHs together with BC in surface sediments from the East Siberian Arctic Shelf (ESAS) were analyzed. Positive matrix factorization (PMF) elucidated a mixed petrogenic and pyrogenic sources and distinct PAH fates associated with diverse input pathways including coastal permafrost erosion contribution ( $\sim$ 30%), petrogenic-related emission ( $\sim$ 34%), fossil fuel combustion (~26%), and biomass burning (~10%). Correlation analysis indicated that BC plays a key role in affecting the behavior and fates of PAHs. In the Chukchi Sea, PAHs are closely associated with soot-BC, whereas in the Laptev Sea (LS) and west East Siberian Sea (W-ESS), they exhibit a coupling process with char-BC. The presence of these carbonaceous materials in the sediments of CS is likely influenced by atmospheric deposition and biological activity, whereas the LS and W-ESS regions are mainly affected by long-distance river transport and direct deposition from coastal permafrost. As global warming continues, permafrost thawing induces the remobilization and retranslocation of PAHs, thereby becoming a significant PAH contributor and input pathway in the rapidly changing Arctic coastal margin.

**Plain Language Summary** The Arctic region is experiencing rapid climate changes, such as reducing sea ice, thawing permafrost, and increasing wildfires. These changes are raising the levels of semi-volatile pollutants such as polycyclic aromatic hydrocarbons (PAHs) in the Arctic coastal margins, with the potential for the region to become a significant reservoir of sedimentary PAHs. In our study, we examined the large-scale distribution and fates of PAHs and black carbon (BC) across the East Siberian Arctic Shelf (ESAS), the world's largest shelf system. We discovered that PAHs have mixed sources, including both petrogenic and pyrogenic origins. We also observed differences in how PAHs were transported and sequestrated in coastal versus open sea areas. In the western ESAS, PAHs and BC are mainly influenced by terrestrial inputs such as river discharge and coastal permafrost erosion. In contrast, in the eastern open-sea regions, they are primarily constrained by atmospheric transport and biological activity are the primary influencing factors. The significant emissions from thawing permafrost along the coasts highlight the growing importance of permafrost-related erosion as a significant contributor of PAHs in the Arctic Ocean, particularly in the context of future warming.

# 1. Introduction

Recently, warming has been accelerating faster in the Arctic than in other parts of the world (Rantanen et al., 2022), potentially making the region the most vulnerable to climate change (Meredith et al., 2019; Terhaar et al., 2021). This vulnerability is characterized by increasing coastal permafrost erosion (Schuur et al., 2022), decreasing sea ice (Olonscheck et al., 2019) and consequent enhancement of ocean primary productivity (Arrigo et al., 2012). Furthermore, as permafrost continues to degrade, wildfires occur more frequently in boreal forests



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Writing – original draft: Wenxiu Yu, Limin Hu and tundra across Siberian ecosystems (Tomshin & Solovyev, 2022), releasing large amounts of pyrogenic carbon and permafrost carbon into the coastal margins (Sparkes et al., 2018). These changes have led to increasing concern about the various climate-related feedback (Talucci et al., 2022). Polycyclic aromatic hydrocarbons (PAHs), a class of semi-volatile persistent organic pollutants (POPs), are typical molecular markers of pyrogenic carbonaceous materials (Balmer et al., 2019; Laender et al., 2011; Zhang et al., 2023). PAHs are typically released into the environment from both pyrogenic and petrogenic sources (Saha et al., 2009; Yunker et al., 2002). They possess the capability to reach the Arctic region via long-range atmospheric or oceanic transport as well as local emissions (MacDonald et al., 2000). Despite reductions in global emissions, PAHs present in the Arctic air have not exhibited a notable downward trend (Yu et al., 2019). In the context of Arctic warming, increasing land-based inputs such as river discharge and coastal erosion, permafrost retreat, and more frequent wildfires further contribute to the supply of PAHs in the Arctic (Hung et al., 2022; Song et al., 2023; Yunker et al., 2011). Moreover, the rapid loss of sea ice coupled with enhanced biological pump process could facilitate PAHs sinking through the water column, promoting their accumulation in bottom sediments (Dai et al., 2019; Galbán-Malagón et al., 2012; Ma et al., 2011). These processes profoundly impact the biogeochemical behaviors of PAHs in polar environments.

After their release into the environment, PAHs exhibit strong hydrophobicity and corresponding affinity for particles (Accardi-Dey & Gschwend, 2002; Cornelissen et al., 2005). Previous research studies have shown that particle sinking and export are the primary mechanisms for removing PAHs from Arctic seawater, whether these PAHs from air–sea exchange, dry and wet deposition, riverine input, or permafrost thaw (Ke et al., 2017; Lin et al., 2022; Liu et al., 2021). Therefore, marine sediments serve as the ultimate sink for these particle-reactive organic compounds (Suman et al., 1997). Previous studies have revealed that PAH compounds are absorbed into amorphous organic matter (Huang et al., 1997; Lambert, 1968). In many situations, they also tend to be strongly adsorbed onto condensed carbonaceous materials such as black carbon (BC) (Cornelissen et al., 2005). BC, a byproduct of either the combustion of biomass (known as char) or fossil fuels (commonly termed soot), is an important carbonaceous sorbent (Lohmann et al., 2005; Schmidt & Noack, 2000). Many studies have revealed associations between PAHs and BC, particularly in polar environments such as the Swedish shelf and Pan-Arctic rivers (Elmquist et al., 2008; Sánchez-García et al., 2010), emphasizing the constraint effects of pyrogenic BC on PAH occurrences. The adsorption capacity of the BC fraction can constraint the PAHs occurrences and preservation in sediments, thereby helping to elucidate the behaviors and fates of PAHs in various environmental matrices (Jonker & Koelmans, 2002).

The East Siberian Arctic Shelf (ESAS) is a highly vulnerable region, characterized by intense sea ice melting, permafrost degradation, and altered vegetation in the context of rapid Arctic climate change (Thomas et al., 2022); these changes can significantly impact the behaviors and fates of carbonaceous materials. Constrained by limited land-based input due to the absence of large-scale rivers and high primary productivity within the eastern ESAS (i.e., the Chukchi Sea) (Viscosi-Shirley et al., 2003), long-range atmospheric delivery and biological processes (biodegradation, adsorption, and biological pump) can significantly affect the deposition of hydrophobic PAHs (González-Gava et al., 2019; Liu et al., 2021; Nizzetto et al., 2012). Conversely, in the western ESAS, terrestrial inputs from river discharge and coastal permafrost erosion are important sources of PAHs and BC (Petrova et al., 2008; Salvadó et al., 2017; Vonk et al., 2012; Yunker et al., 2011). Permafrost thaw slumping has driven PAH compounds into the sediments, as reported in the Mackenzie Delta (Thienpont et al., 2020). However, few studies have been conducted on the Eurasian Arctic margins, which were previously poorly recognized but maybe an important source of remobilized PAHs (Lin et al., 2022). The actual export of these PAHs from coastal permafrost, as well as their composition and fate on the adjacent Siberian Arctic shelf, remains unclear. There is only limited evidence that petrogenic hydrocarbons in the central Arctic Ocean are associated with substantial eroded coal transport from the Lena River watershed or the Laptev Sea (LS) coast (Boucsein et al., 2002; Yunker et al., 2011). Consequently, explorations of PAH spatial heterogeneity, sources, and coastal fates, as well as the role of BC in determining the transport and fate of PAHs, are important for identifying the source and sink processes of these carbonaceous components in a region that experiences some of the most intense land-sea interactions globally.

In this work, we examined the occurrences and biogeochemical behaviors of PAHs and two subtypes of BC in sediments from the ESAS. We quantitatively analyzed their spatial heterogeneity and coastal fates and linked these results to biological inputs and permafrost erosion processes across the broad and shallow Arctic coastal margins.





**Figure 1.** Map showing the study area in the Arctic Ocean and the East Siberian Arctic Shelf, with rivers marked in blue and ICD in green. Coastal erosion rates are displayed by yellow, orange, and red lines (Lantuit et al., 2012). Black arrows represent typical currents, following Tesi et al. (2016): BG, Beaufort Gyre; TPD, transpolar drift; PI, Pacific inflow; SCC, Siberian Coastal Current. Arctic lignite and bituminous coal deposits are based on the 1978 Polar Regions Atlas (Yunker et al., 2011). Wildfire locations in major Siberian watersheds from 2016 to 2018 are depicted as red dots (https://firms.modaps.eosdis.nasa.gov/).

# 2. Materials and Methods

#### 2.1. Regional Setting and Sampling

The ESAS is one of the largest shelf systems in the world, spanning up to 800 km wide with a mean depth of 50 m (Stein & Macdonald, 2004). A primary feature of coastal regions along the ESAS is permafrost, typically consisting of late Pleistocene ice complex deposits (ICD, also known as Yedoma) (Figure 1) (Günther et al., 2013; Lantuit et al., 2013). The ESAS receives extensive terrestrial carbon inputs from rivers and permafrost thawing; in particular, riverine and coastal erosion release a large amount of old carbon from permafrost (Günther et al., 2013; Vonk et al., 2012). The LS receives large amounts of freshwater (approximately 745 km<sup>3</sup> yr<sup>-1</sup>) mainly transported by the Lena River (Semiletov et al., 2000), although most carbon inputs arise from coastal erosion of ICD permafrost (Semiletov et al., 2012; Vonk et al., 2012). Enhanced sea ice melting and freshwater input via the Pacific inflow (PI) stimulate phytoplankton productivity, making the Chukchi Sea (CS) one of the most productive regions in the Arctic marginal seas (Payne et al., 2021). The East Siberian Sea (ESS) consists of two contrasting physical and biogeochemical regimes. The eastern ESS (E-ESS, 160°E–180°E) is influenced by the PI, resulting in relatively high primary productivity (similar to productivity in the CS), whereas the western ESS (W-ESS, 140°E–160°E) is controlled by riverine inputs and severe coastal erosion at a rate of 3–4 m yr<sup>-1</sup>; the erosion-prone ICD permafrost serves as the main sediment source (Lantuit et al., 2012; Semiletov et al., 2005).

The locations of sediment sampling sites in the ESAS are illustrated in Figure 1. In total, 74 surface sediment samples (0–1 cm depth) were collected using a stainless-steel box corer during joint cruises in 2016 (Lv77) and 2018 (Lv83) onboard *R/V* Akademik M.A. Lavrentiev. Sediment samples were wrapped in aluminum foil and stored at  $-20^{\circ}$ C until analysis.

# 2.2. Analytical Procedures

The procedure for PAH analysis, as well as the quality assurance and quality control (QA/QC) protocols, followed the methods described by Mai et al. (2003) and Lin et al. (2011). Briefly, homogenized sediment samples (approximately 10 g each) were first spiked with a mixture of five deuterated PAHs (naphthalene-d<sub>8</sub>, acenaphthene-d<sub>10</sub>, phenanthrene-d<sub>10</sub>, acenaphthene-d<sub>12</sub>, and perylene-d<sub>12</sub>) recovery surrogate standards, and then subjected to ultrasound extraction with dichloromethane. The extracts were concentrated and fractionated using a silica-alumina (1:1) column. The target compounds are the 16 priority PAHs proposed by the U.S. EPA including naphthalene (Nap), acenaphthylene (Ac), acenaphthene (Ace), fluorene (Flu), phenanthrene (Phe), anthracene (Ant), fluoranthene (Fluo), pyrene (Pyr), benz[*a*]anthracene (BaA), chrysene (Chr), benzo[*b*]fluoranthene (BbF), benzo[*k*]fluoranthene (BkF), benzo[*a*]pyrene (BaP), indeno[1,2,3-*cd*]pyrene (InP), dibenz[*a*,*h*]anthracene (DBA), and benzo [*ghi*]perylene (BghiP), along with methylphenanthrene (M-Phe), perylene and retene. Hexamethylbenzene was added as an internal standard. Each mixture was reduced and subjected to analysis via gas chromatography with a mass-selective detector (Agilent series 7890B mass spectrometer interfaced with a 5,977 series gas chromatograph) equipped with an HP-5 capillary column (25 m × 0.25 mm inner diameter; film thickness, 0.25 µm).

For QA/QC, procedural blanks, standard-spiked blanks, standard-spiked matrices, and duplicate samples were analyzed simultaneously. The limits of detection for individual PAHs ranged from 0.2 to 2 ng/g for 10 g sediment samples (Mai et al., 2003). Procedural blanks contained no detectable amounts of the target PAH compounds. The PAH recoveries in the standard-spiked matrices ranged from 78% to 93%; paired duplicate samples demonstrated agreement to within 15% of the measured values (-10). The mean surrogate recoveries were 65 ± 11% for naphthalene-d<sub>8</sub>, 60 ± 6% for acenaphthene-d<sub>10</sub>, 98 ± 14% for phenanthrene-d<sub>10</sub>, 102 ± 14% for chrysene-d<sub>12</sub>, and 99 ± 10% for perylene-d<sub>12</sub>. Concentrations reported here were not corrected for recovery.

For BC analysis, wet-chemical pretreatment combined with the thermal optical reflectance (TOR) detection method of Han, Cao, Chow, et al. (2007) was adopted. Briefly, thawed, freeze-dried, and homogenized (<80 mesh) sediment samples were acid-treated to effectively remove carbonates, silicates, and secondary minerals; the residues were then filtered through precombusted quartz fiber filters to achieve an even distribution. The filters were analyzed for BC on a DRI Model 2001 thermal/optical carbon analyzer in accordance with the IMPROVE\_A protocols. Detailed BC analytical procedures were described by Fang et al. (2015). For QA/QC, 10% of filters were randomly selected with two punches per filter analyzed for BC. The measured BC concentration had a relative standard deviation (RSD) ranging from 1% to 10%, averaging 4%, indicating an even distribution. Blank and replicate samples and standard reference material (NIST SRM-1941b) were analyzed at a rate of one per 10 samples. Blank samples (n = 19) yielded  $0.0 \pm 0.0 \,\mu$ g/cm<sup>2</sup> for BC, and the RSD for 19 replicates ranged 0%–13%, averaging within 4%. The measured BC concentration in SRM-1941b was 10.6 ± 1.9 mg/g (n = 19), which was well in accordance with the reported values (Cong et al., 2013; Hammes et al., 2007; Han, Cao, An, et al., 2007). More detailed procedures regarding the QA/QC of BC are available in Text S1 of Supporting Information S1.

The analytical methods of Hu et al. (2009) were utilized for total organic carbon (TOC) and grain size. Replicate analysis of one sample (n = 6) provided a precision of ±0.05 wt.% for TOC. For grain size, the relative error of the duplicate samples was <3% (n = 6).

#### 2.3. Principal Component Analysis (PCA)

PCA, a multivariate analytical tool, was used to determine sample distributions and assess relationships among measured parameters. Prior to analysis, non-detectable values were replaced with concentrations equal to one-half of the method detection limits. PCA was performed using Origin 2023b (OriginLab Corporation) to extract the principal components (PCs) based on the correlation matrix.

#### 2.4. Positive Matrix Factorization (PMF) Modeling

PMF is a PCA-based receptor model with non-negative constraints that involves the solution of quantitative source apportionment equations with oblique solutions in a reduced dimensional space. The detailed concept and application of PMF source apportionment are described in the EPA PMF 5.0 Fundamentals and User Guide (www.epa.gov/heasd/products/pmf). In principle, the PMF model is based on the following equation,



$$Xij = \sum_{k=1}^{p} AikFkj + Rij$$

where *Xij* is the concentration of the *j*th congener in the *i*th sample of the original data set; *Aik* is the contribution of the *k*th factor to the *i*th sample; *Fkj* is the fraction of the *k*th factor arising from congener *j*; and *Rij* is the residual between the measured *Xij* and the predicted *Xij* using principal components.

$$\mathbf{Q} = \sum_{i=1}^{n} \sum_{j=1}^{m} \left( \frac{Xij - \sum_{k=1}^{p} AikFkj}{Sij} \right)^{2}$$

where Sij is the uncertainty of the *j*th congener in the *i*th sample of the original data set containing *m* congeners and *n* samples; Q is the weighted sum of squares of differences between the PMF output and the original data set. One of the tasks of PMF analysis is minimizing the Q value.

Before PMF analysis, the concentration file and corresponding uncertainty file were input into the model. In this study, an uncertainty value of 20% for PAHs was adopted based on analyses of replicate samples and standard reference material (Mai et al., 2003). In this study, the input data for PMF analysis were 13 PAHs results from 74 sediment samples. Because Nap, Ac, Ant, and DBA were lower than their method detection limits (MDL) in the majority of samples, they were excluded from the initial PMF analysis. The 4-factor solution produced a Q value of 897.4, which is very close to the theoretical value of 923.4 (Figure S1 in Supporting Information S1).

# 3. Results and Discussion

#### 3.1. Occurrences and Spatial Variations of PAHs and BC in the ESAS

The total concentrations of 16 PAHs in ESAS sediments ranged from 3.9 to 69 ng/g dry weight (dw), with a mean of 27.6 ng/g dw. The PAH composition was dominated by low-molecular-weight (LMW) PAHs, which explained 64.7% of the total variance, with the Phe being the primary component. The concentrations of M-Phe in the ESAS sediments ranged from 1 to 42 ng/g dw, exhibiting a spatial heterogeneity, with higher abundance observed in both nearshore and open sea area (Figure S2 in Supporting Information S1). The BC content ranged from 0.1 to 2.3 mg/g dw (with a mean of 1 mg/g dw), corresponding to 15%–40% of TOC; char-BC accounted for more than 70% of the total BC. The abundances of PAH components were substantially lower than observations in other Arctic region sediments, such as those of the Barents Sea (2,109 ± 1,640 ng/g dw) and Svalbard inshore region (8,967 ± 5,167 ng/g dw) (Dahle et al., 2006), probably because of proximity to continental sources and the inclusion of alkyl-PAHs in  $\Sigma$ PAHs. Moreover, PAH contents in this work were comparable to or slightly lower than previous results in the LS and ESS (38–223 ng/g dw), CS (52.4–91.3 ng/g dw), and five Siberian rivers (23.8–129 ng/g dw) (Chen et al., 2018; Elmquist et al., 2008; Lakhmanov et al., 2022).

Spatially, the highest PAH concentrations were observed in the CS and E-ESS shelf break regions, whereas lower levels were present around the New Siberian Islands and the Kolyma River mouth (Figure 2a). Lin et al. (2020) observed a sharp increase in the concentration of PAHs in the northern CS, particularly in the vicinity of the shelf breaks in the Beaufort Sea. Additionally, high PAH levels along the marginal E-ESS shelf break align with the distribution of the ice edge as delimited by  $IP_{25}$  (ice proxy lipid biomarker with 25 carbons) and brassicasterol records (Figure 2a). High concentrations of brassicasterol are present in the offshore area around 74°N of the ESS with a sharp decrease in the northern ESS (Figure S3 in Supporting Information S1; Su et al., 2022). Therefore, biological processes in these regions with high settling efficiency may be the main drivers of the accumulation and precipitation of hydrophobic, lipophilic PAHs into sediments (Liu et al., 2021; Nizzetto et al., 2012; Wan et al., 2022). Moreover, the increase in primary production associated with recent warming may exert further constraints on PAH behavior in the Siberian Arctic region.

In addition to biological influences, the sedimentation regime and  $\delta^{13}$ C values indicated dominant terrigenous inputs in the western coastal ESAS driven by riverine runoff and coastal permafrost erosion (Günther et al., 2013; Myers-Pigg et al., 2015; Vonk et al., 2012). The highest BC content and a relatively high concentration of PAHs were observed at the mouths of the Lena and Yana Rivers, with prominent decreases were evident on the outer shelf, suggesting a significant effect of runoff input (Elmquist et al., 2008; Petrova et al., 2008). It has been



**Figure 2.** Spatial distributions of 16 PAHs (a), LMW PAHs (b), high-molecular-weight (HMW) PAHs (c), BC (d), char (e), soot (f), BC/TOC (g), and char/soot (h) in the ESAS. LMW: sum of 2- and 3-ring PAHs, HMW: sum of 4-, 5-, and 6-ring PAHs.

reported that the prevailing cold climate has helped the preservation and accumulation of PAHs and other POPs (e.g., polychlorinated biphenyls) in permafrost (Ren et al., 2019; Thienpont et al., 2020; Wei et al., 2024; Zhu et al., 2023). This indicates that the widespread permafrost in Siberian Arctic margins could be an important potential reservoir of PAHs and BC (Guo et al., 2004; Salvadó et al., 2017).

# 3.2. Composition and Sources Identification of PAHs

# **3.2.1. Diagnostic Ratios**

Diagnostic ratios of PAH species with the same isomeric structure are widely used to determine PAH origins (Yunker et al., 2002). PAH ratios such as Fluo/(Fluo + Pyr) and InP/(InP + BghiP) from this study, along with





**Figure 3.** Ratios of InP/(InP + BghiP) versus Fluo/(Fluo + Pyr) in the ESAS. Black and red ellipses represent nearshore and outer shelf samples, respectively. Diamonds and triangles represent the stations of Lin et al. (2020) and Lakhmanov et al. (2022), respectively.

previous literature, are presented in Figure 3. These findings suggested a mixed contribution of pyrogenic and petrogenic inputs (Lakhmanov et al., 2022; Lin et al., 2020). In the ESS, increasing trends of both Fluo/(Fluo + Pyr) and InP/(InP + BghiP) ratios were observed as samples moved from nearshore region to the outer shelf (Figure 3). The relatively high proportion of petrogenic PAHs in the nearshore region may be due to land-based terrestrial inputs from rivers and coastal permafrost release, whereas pyrogenic PAHs in the offshore area were mainly derived from long-range atmospheric transport (Lin et al., 2020; Yunker et al., 2011). For example, it has been reported that wildfires in Russia, along with air pollutants from Eurasia transmitted to the Arctic during the cold seasons, have contributed to the levels of pyrogenic PAHs in the Arctic atmosphere (Ding et al., 2007; Hallsal et al., 1997; Kharuk et al., 2022).

# 3.2.2. Composition and Sources of PAHs Based on PCA

The PAHs present in the Arctic shelf sediments have undergone long-range transport via the atmosphere, ocean currents, and rivers (Sofowote et al., 2011; Zhang et al., 2023). Additionally, these transport process could be further modulated by local inputs such as coastal erosion and biological processes (Lin et al., 2022; Liu et al., 2021; Yunker et al., 2011). To examine

the PAH sources and their various input pathways within this large-scale shelf system, monomer PAH compounds (including perylene), as well as parameters such as char, soot, TOC, and opal, were conducted for PCA analysis. As shown in Figure 4, three different clusters were identified based on the first three PC scores of each sample, representing distinct input pathways. The first three PCs (PC1, PC2, and PC3) explained 43.9%, 14.9%, and 11.0% of the total variance, respectively. PC1 was heavily weighted by 4-ring PAHs (such as Chr, BaA, Fluo, and Pyr), with moderate loading for 5 + 6 ring PAHs (Figure 4a), suggesting a pyrogenic origin. Samples from the CS exhibited high PC1 loadings, and these HMW PAH components likely originated from atmospheric transport and deposition (Ma et al., 2017; Sofowote et al., 2011). PC2 was highly loaded in perylene, char, soot, and TOC, particularly corresponding to the high scores observed in samples from the western ESAS (e.g., the LS) (Figures 4a and 4b), suggesting a strong input from the riverine discharge. Perylene, a soil-derived terrestrial component formed at the stage of the early diagenesis of biogenic components (Hanke et al., 2019), dominated the sediments in the Lena River estuarine and coastal areas (Figure S5a in Supporting Information S1), indicating a direct influence from fluvial input (Petrova et al., 2008). Moreover, a significant positive correlation between BC and TOC ( $r^2 = 0.87$ , p < 0.001) was also observed in the Laptev Sea, accompanied by a deficit of  $\delta^{13}$ C occurrence,



Figure 4. Loadings of the PCA model of 16 PAHs and other parameters and corresponding sample scores of three PCs.





**Figure 5.** Comparisons of PMF model derived four-factor profiles with published results: (a) permafrost-related input (Vasil'chuk et al., 2020); (b) oil and gas activities (Ma et al., 2017, 2020); (c) fossil fuel combustion (Sofowote et al., 2011); (d) biomass burning (Sofowote et al., 2011).

further emphasizing the dominance of land-based input (Figures S5 and S6 in Supporting Information S1; Vonk et al., 2012; Günther et al., 2013). PC3 was strongly correlated with LMW PAH homologs, such as Flu, Ace, Phe, and Ac (Figure 4b). Notably, samples from W-ESS showed relatively high PC3 scores, potentially indicating a generic petrogenic contribution related to coastal permafrost erosion (Thienpont et al., 2020; Yunker et al., 2011; Vasil'chuk et al., 2020). Besides, in addition to the high presence of M-Phe in the nearshore area (Figure S2 in Supporting Information S1), their high abundance in the open sea area may also be associated with the biological-related process (Ma et al., 2020).

#### 3.3. Evaluation of the Coastal Fates of PAHs in the ESAS Through PMF Modeling

#### 3.3.1. PMF Modeling

PMF can quantitatively estimate the relative contributions of PAHs from specific source categories (Stout & Graan, 2010). After the testing of 3–7 factors, a four-factor solution was adopted based on comparisons with reported PAH source profiles within the Arctic (Lin et al., 2020; Vasil'chuk et al., 2020; Ji et al., 2019; Ma et al., 2017; Sofowote et al., 2011). The correlation between the estimated and measured concentrations was almost unity, suggesting that the measured concentrations were adequately explained by these four factors.

Factors 1 and 2 were dominated by LMW PAHs, especially Phe and Flu (Figures 5a and 5b); this petrogenic profile in the Arctic environment may be related to oil and gas activities (AMAP, 2010; Harvey et al., 2014; Ma et al., 2017; Yunker & MacDonald, 1995) and/or the contributions of eroded source rocks rich in organic matter and coal deposits (Fahl et al., 2003; Lin et al., 2020; Yunker et al., 2011). The profile of factor 3 was consistent with generic fossil fuel combustion emissions (including coal, oil, and gasoline engines) (Liu et al., 2003; Sofowote et al., 2011; Yunker et al., 2002), characterized by high levels of BaA, Chr, BbF, BkF, InP, and BghiP with moderate loading for Fluo (Figure 5c). Factor 4 was identified as biomass burning due to the high loading of





Figure 6. Spatial patterns and contributions of four PMF-resolved factors to PAH abundance in each sediment sample from the ESAS. Ellipses represent the proportional contributions of each factor in the coastal sites of the LS and W-ESS, outer shelf sites of the ESS, and the entire CS.

Pyr (Figure 5d), which serves as a marker of wood or grass combustion (Sofowote et al., 2011; Yu et al., 2019). Nevertheless, considering that Pyr could originate from various sources, retene might serve as a better marker in this context (Figure S7 in Supporting Information S1; Ramdahl, 1983).

# 3.3.2. Implications for the Input Pathways and Coastal Fates of PAHs in the ESAS

The spatial patterns of various PAH profiles in the ESAS are presented in Figure 6. Factor 1 exhibited the relatively high concentrations in the nearshore areas of the LS and W-ESS, which could be subject to the influences of permafrost erosion and riverine input (Stein & Macdonald, 2004; Vonk et al., 2012). Moreover, based on comparison with reference source profiles of permafrost-affected soils (Vasil'chuk et al., 2020; Thienpont et al., 2020; Pastukhov et al., 2021; Wei et al., 2024), the estimated contribution of factor 1 accounting 30% of the total variance could be likely linked to permafrost-related emissions. This indicated a potential input pathway for these petrogenic PAH components in the ESAS (Lin et al., 2022; Thienpont et al., 2020). Due to the input of substantial amounts of terrigenous material from the Lena River, which drains a watershed primarily composed of continuous permafrost (Figure 1) (Miner et al., 2022), PAHs with highly petrogenic signals in the Lena River estuary presumably have undergone long-distance transport via rivers (Yunker et al., 2011). As warming intensifies, enhanced riverbank erosion and elevated runoff (Tananaev & Lotsari, 2022) have possibly resulted in the migration of PAHs from watershed permafrost into riverine systems (Elmquist et al., 2008; Jin et al., 2022), which ultimately deposited in the adjacent Arctic shelf. Previous studies have found that petrogenic hydrocarbons in sediments of the central Arctic Ocean were associated with substantial eroded coal transported from the Lena River watershed (Yamamoto et al., 2008; Yunker et al., 2009, 2011). Moreover, the release of PAHs from coastal erosion in the W-ESS is a rapid and abrupt collapse process (Turetsky et al., 2019). This coastal permafrost slumping could introduce previously bound PAHs from the terrestrial geology to the shelf ecosystem, which are shown to be more indicative of petrogenic sources (Thienpont et al., 2020).

In addition to land-based input, atmospheric transport and subsequent deposition also represent a significant PAH contribution within the Arctic coastal shelf system (Ding et al., 2007; Halsall et al., 1997; Ma et al., 2013). PMF analysis revealed that atmospheric source accounts for a higher proportion of PAHs in open-sea regions (e.g., outer shelf sites and CS) than those in the coastal margins (Figure 6), indicating that atmospheric deposition represents a primary input pathway in the ESAS. Biomass burning constitutes another prominent portion of the atmospheric load, and mostly originating from numerous circum-Arctic vegetation fires, particularly in the most fire-prone areas of the Siberian Arctic (Kharuk et al., 2022; Soja et al., 2004). It was reported that atmospheric circulation can transport wildfire smoke, carrying PAH compounds and carbon components, such as soot, northward to the Arctic (Kim et al., 2005; Song et al., 2023).

Due to the lipophilicity of PAHs, the influences of biological activity on PAH settlement in the highly productive Chukchi Shelf cannot be neglected (Galbán-Malagón et al., 2012; Liu et al., 2021). HMW PAHs are more likely to bind to particles produced by organisms and settle vertically into sediments, whereas LMW PAHs, which are more water-soluble, are easily taken up by plankton and accumulated in their bodies (González-Gaya et al., 2019; Nizzetto et al., 2012). The concentrations of LMW PAHs are highest in samples from the highly productive CS





\*p<=0.05 \*\*p<=0.01 \*\*\*p<=0.001



(Figure 6), where 38% of total primary productivity is contributed by the phytoplankton ( $<2 \mu m$ ) such as diatoms (Park et al., 2022). Furthermore, this PAH profile demonstrates a resemblance to the reported compositions of plankton and benthos samples, which were dominated by LMW PAHs (e.g., Phe, Ace, Flu, and Fluo) (González-Gaya et al., 2019; Ma et al., 2020). Previous studies have found that two diatoms, *Nitzschia sp.* and *Skeletonema costatum*, can accumulate Phe and Fluo (Hong et al., 2008), thereby representing a potential biogenic-related constraint on the occurrence of these LMW PAHs. With continued warming, increased air-sea exchange, biotic degradation, and photolytic degradation are regarded as important factors affecting PAH behaviors. Together with the biological processes, these factors jointly regulate the sources and fates of PAHs (González-Gaya et al., 2016; Ke et al., 2017; Ma et al., 2011).

#### 3.4. Correlation Between the PAHs and BC and Their Environmental Implications

BC shares similar sources with pyrogenic PAHs (primarily composed of HMW PAHs) from incomplete combustion processes (Gustafsson & Gschwend, 1997; Jonker & Koelmans, 2002; Lohmann et al., 2005), and its strong adsorption capacity significantly impacts the occurrence of sedimentary PAHs (Accardi-Dey & Gschwend, 2002; Jonker & Koelmans, 2002). Based on the results of PMF source apportionment (Figure 6), we performed a correlation analysis between BC and PAHs in the CS, where atmospheric deposition was the primarily influencing factor, and compared it to that in the LS and W-ESS, which were mainly affected by terrestrial input (Figure 7).

In the present study, a significant correlation (r = 0.71, p < 0.05) was observed between HMW PAHs and soot-BC, but only in the CS (Figure 7a). This subtype of BC is the smaller combustion particulates (0.1–1 µm) which are readily transported and deposited via atmosphere (Gustafsson et al., 2001; Wang et al., 2013). The associations between soot particles and HMW PAH compounds are attributable to two reasons. First, soot-BC has a strong affinity for nonpolar substances such as PAHs (Jonker & Koelmans, 2002). Second, HMW PAHs, as molecular precursor of soot (Richter & Howard, 2000), can be co-emitted with soot into the atmosphere, making them likely to be co-deposited to sediments (Figure 8; Han et al., 2015). Therefore, the co-occurrence of HMW PAHs and soot-BC indicated similar input pathways (e.g., atmospheric loading) for these pyrogenic particulates on the CS shelf (Wang et al., 2013). In addition, the biological processes serve as an alternative input mechanism and play a dominant role in PAH behaviors in this high-productivity area (Figure 8; Galbán-Malagón et al., 2012; Berrojalbiz et al., 2011). Due to the recent thinning of sea ice and the substantial increase in light transmittance within Arctic waters (Ardyna et al., 2022; Arrigo et al., 2012), the biological constraint is receiving increasing attention regarding its role in modulating atmospheric transport and the fate of PAHs.

In contrast, the western ESAS is affected by both river discharge and ice-rich permafrost erosion, which could release the PAHs and carbonaceous materials stored in watershed and coastal permafrost (Sparkes et al., 2018; Yunker et al., 2009, 2011). The large Arctic rivers (e.g., the Lena, Indigirka, and Kolyma Rivers) mostly drain





**Figure 8.** Schematic illustration of PAHs and BC input and deposition heterogeneity in the Arctic coastal margin. Pyrogenic PAHs, originating from fossil fuel combustion and wildfires, are transported via the atmosphere. They settle either by combining with soot-BC particles or through biological deposition in open-sea regions (Ma et al., 2020; Wang et al., 2013). As for terrestrial inputs, rivers delivered char-BC and pyrogenic PAHs into the adjacent Siberian shelf regions. Coastal and watershed permafrost erosion primarily contribute to petrogenic PAHs (Yunker et al., 2011).

catchments containing permafrost and vegetation fires, such as southern Siberian taiga and tundra wildfires (Figure 1; Talucci et al., 2022). Unlike in the CS, char-BC and TOC are well correlated with PAHs in the LS and W-ESS (Figure 7b). This BC fraction consists of larger particle size compared to atmospheric soot and tends to deposit in close to the source area (Schmidt & Noack, 2000). The partial combustion-derived PAHs emitted into the atmosphere could settle in rivers or onto the topsoil-permafrost, and subsequently be captured by these carbonaceous particles (e.g., char-BC and TOC) (Nam et al., 2008). As the permafrost thaws, they could be delivered together into adjacent Siberian shelf regions through fluvial transport (Figure 8; Adhikari et al., 2019; Masiello & Louchouarn, 2013; Myers-Pigg et al., 2015). Moreover, the PAHs correlate more closely with char-BC than TOC, which could be attributed to the higher sorption capacity of this carbon faction (Han et al., 2015). Char-BC has high internal microporosity, providing a solid matrix that traps and stabilizes PAHs (Bornemann et al., 2007; Yunker et al., 2011). Therefore, the fates of PAHs in the western shelf could be largely constrained by char-BC through co-transport processes, and/or post-depositional particle bonding (Yunker et al., 2011). As the permafrost input become a potentially important contributor of PAHs with climate warming, the closer associations indicate that the terrestrial carbonaceous particles would play a growing role in determining the coastal fates of PAHs in Arctic coastal margins.

# 4. Conclusions

The occurrence of PAHs in the ESAS sediments was dominated by LMW PAHs, with Phe being the main component. Significant spatial variations of PAHs concentration were observed across the ESAS, with the highest level observed in the CS and at the ice edge along the eastern ESAS break. PAHs exhibited a mixture of petrogenic and pyrogenic sources, with distinct fates related to diverse input pathways in coastal and open sea areas of the ESAS. The deposition and burial of pyrogenic PAH compounds originating from fossil fuel and biomass combustion, were significantly enhanced by biological processes, particularly on the Chukchi Shelf. In addition, BC played a key role in affecting the behavior and fates of pyrogenic PAHs. Specifically, PAHs exhibited a close association with soot-BC in the CS, but shared coupled processes with char-BC in the western ESAS. The petrogenic PAHs identified through the PMF model displayed varied input mechanisms, which explained a regional disparity in PAHs sequestration. These LMW PAHs in the sediments of CS could be more linked to plankton uptake and sinking, whereas their presence in the LS and W-ESS should be significantly influenced by the land-based processes (e.g., fluvial discharge and coastal permafrost erosion). The identification of the significant signals of permafrost-related PAHs emissions in the Siberian coastal area underscored the concern that



permafrost erosion could become a significant contributor and influencing factor in determining the coastal fate of these carbonaceous materials in Arctic environments under future warming scenarios.

#### **Data Availability Statement**

The original data in present are available in Yu et al. (2024). The Sea-ice concentrations (SIC) data from 2007 to 2016 are available from the National Snow and Ice Data Center (NSIDC, https://nsidc.org). The wildfires data detected with Moderate Resolution Imaging Spectroradiometer (MODIS) wildfire data are obtained from the Fire Information for Resource Management System (FIRMS, https://firms.modaps.eosdis.nasa.gov/). The detailed concept and application of PMF source apportionment are described in the EPA PMF 5.0 Fundamentals and User Guide (www.epa.gov/heasd/products/pmf).

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