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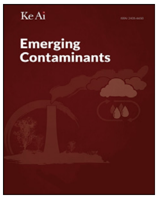

Sources and environmental fate of pyrogenic polycyclic aromatic hydrocarbons (PAHs) in the Arctic

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ABSTRACT

Polycyclic aromatic hydrocarbons (PAHs) are large class of hydrophobic, semi-volatile organic contaminants that may enter the environment from both natural sources and anthropogenic activities. Pyrogenic PAHs arise from the incomplete combustion of fossil fuels and organic matter and following dispersal via long-range transport and may subsequently deposit in surface waters, soils and sediments of remote regions, including the Arctic. The current review summarizes and discusses Arctic data that is available for combustion-derived PAHs between 2004 and early 2018, focusing largely on data collected from remote, unexploited Arctic regions and from studies that provide some evidence of a pyrogenic origin. The increasing use of attribution ratios, which aid in discriminating PAHs from petrogenic or pyrogenic sources, suggest PAHs found in Arctic marine waters and sediment predominantly originate from natural underwater seeps, while those measured in air, freshwater, and terrestrial environments are likely to have originated from atmospheric and combustion-derived sources. Modeling efforts indicate that atmospheric PAHs in the Canadian and Norwegian Arctic are likely to have originated in the northern hemisphere – predominantly from Western Russia, northern Europe, and North America. East Asia appears to be a minor source of PAHs to the Arctic, despite contributing more than 50% of global PAH emissions. In comparison to the growing data for atmospheric PAHs, environmental data for these compounds in terrestrial and freshwater environments remain scarce. PAHs have been detected in Arctic biota from terrestrial, freshwater and marine environments, indicating exposure, however, levels are generally low, as most organisms efficiently metabolize parent PAHs. Globally, PAH emissions are expected to decline in the future, however models suggest the Arctic may not experience the same magnitude of decline projected for other world regions. Furthermore, future changes in climate may contribute to a re-volatilization of environmental PAHs, providing a source of secondary emissions to the Arctic atmosphere, emphasizing the importance of future monitoring for understanding the sources, fate and impacts of PAHs in the Arctic.

1. Introduction

Polycyclic aromatic compounds (PACs) are comprised of several thousand individual compounds, including the better-known subgroup of polycyclic aromatic hydrocarbons (PAHs) and the heterocyclic aromatic compounds. PAHs are hydrophobic, semi-volatile organic contaminants comprising at least two fused aromatic rings and contain only carbon and hydrogen (Fig. 1). Both natural and anthropogenic activities, including coal and petroleum use, vehicle emissions, forest fires and volcanic eruptions, are responsible for the widespread environmental occurrence of PAHs [1,2]. Once

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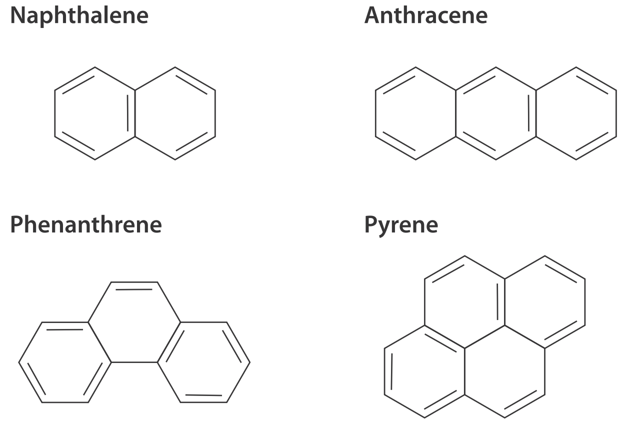

Fig. 1. Four representative polycyclic aromatic hydrocarbons (PAHs). Source: AMAP [14].

released to the atmosphere, PAHs disperse via long-range transport and subsequently deposit in surface waters, soils and sediments of remote regions, including the Arctic [3,4].

Anthropogenic releases, persistence, and toxicity, prompted the designation of PAHs as persistent organic pollutants (POPs) by the UNECE Convention on Long-range Transboundary Air Pollution (LRTAP) and the listing of PAHs by the OSPAR convention [5,6]. While there are several hundred individual PAHs, 16 were selected in the mid-1970s as priority pollutants by the United States Environmental Protection Agency (USEPA) based on their toxicity, ability to be analyzed, and environmental occurrence at the time [7]. These 16 PAHs (Table 1) are frequently targeted for monitoring and have become a *de facto* standard globally [8].

The USEPA priority PAHs tend to predominate in environmental mixtures from pyrogenic sources, i.e. those arising from the incomplete combustion of fossil fuels and organic matter [1,9–11]. Whereas non-combusted fossil fuels contain a high abundance of alkyl-substituted PAHs, the high temperatures of combustion eliminate alkyl side chains, resulting in a higher prevalence of unsubstituted or 'parent' PAHs. Source attribution ratios, which relate the concentration of alkylated-to-parent PAHs, are frequently used to attribute PAHs in environmental samples to either a petrogenic or pyrogenic source [1]. In general, those from pyrogenic sources are dominated by parent PAHs and also contain more of the high-molecular weight parent PAHs. Heterocyclic aromatic compounds (NSO-PACs) – such as those with sulfur (thiophenes), nitrogen (azaarenes) or oxygen (furans) in the ring – are generally present to a lesser degree in pyrogenic materials compared to crude oil and coal-derived materials. Alkylated PAH patterns vary according to origin, with pyrogenic sources typically showing a sloped distribution of $C_0>C_1>C_2>C_3>C_4$ and petrogenic sources showing a bell-shape distribution of C_1–C_4-alkylated PAHs (Andersson and Achten, 2015). Combustion-derived PAHs emitted to the atmosphere are subject to photodegradation, thus PAH transformation products with nitro-, oxy- and hydroxy-substituents may also account for a significant fraction of the total PAHs in air [11].

Table 1
The 16 PAHs defined as priority pollutants by the US Environmental Protection Agency.

Name	CAS#	Rings	MW
Naphthalene	91-20-3	2	128.17
Acenaphthylene	208-96-8	3	152.19
Acenaphthene	83-32-9	3	154.21
Fluorene	86-73-7	3	166.22
Phenanthrene	85-01-8	3	178.23
Anthracene	120-12-7	3	178.23
Fluoranthene	206-44-0	4	202.26
Pyrene	129-00-0	4	202.26
Benzo[*a*]anthracene	56-55-3	4	228.29
Chrysene	218-01-9	4	228.29
Benzo[*b*]fluoranthene	205-99-2	5	253.32
Benzo[*k*]fluoranthene	207-08-9	5	253.32
Benzo[*a*]pyrene	50-32-8	5	253.32
Dibenzo[*a,h*]anthracene	215-58-7	6	278.35
Benzo[*g,h,i*]perylene	191-24-2	6	276.34
Indeno[*1,2,3-cd*]pyrene	193-39-5	6	276.34

PAHs differ from many other POPs and chemicals of emerging concern in that they are unintentional by-products of fossil fuel use and biomass burning and emissions are ongoing. While concentrations of many of the twelve chemicals initially listed under the Stockholm Convention on POPs have declined in Arctic biota over the past 25 years [12], recent modeling studies have suggested that concentrations of PAHs may have risen by as much as 30-fold in fish and mussels, initiating concern over their emerging role in the Arctic ecosystems [13].

This review, originally prepared as part of the recent Arctic Monitoring and Assessment Programme (AMAP) report on 'Contaminants of Emerging Arctic Concern' [14], focuses on PAHs, alkyl-substituted PAHs and degradation products of PAHs, and summarizes the Arctic data available for combustion-derived PAHs between 2004 and early 2018. Because PAHs may originate from sources other than combustion, including natural petroleum seeps and oil and gas activities (the latter addressed in a previous AMAP assessment [15], much of the information used here is taken from reports from remote, unexploited Arctic regions and from studies that provide some evidence that the PAHs were from combustion-derived sources.

1.1. Physical-chemical properties

The general characteristics common among PAHs are high melting and boiling points, low vapor pressures, and very low water solubility which tends to decrease with increasing molecular mass (Table 1). With low vapor pressures, the majority of PAHs, and especially those with three or four rings, are semi-volatile (i.e., compounds with vapor pressures between 10^{-6} and 10^{-2} Pa) and readily undergo long-range transport via the atmosphere. PAHs may be emitted in particulate form or in the gas-phase where they may remain, or sorb to particles such as black carbon, mineral dust and sea salt [16].

Atmospheric PAHs are subject to wet and dry deposition to surface waters and soils, where they have longer lifetimes; models suggest that atmospheric half-lives of 3–5 ring PAHs are of the order of hours or days, whereas their persistence in soils can be on the order of decades [9]. However, their semi-volatile properties allows many PAHs to re-volatilize from terrestrial compartments and surface seawater and re-enter the atmosphere to undergo enhanced long-range transport potential via multi-hopping [16,17].

Although most PAHs are sufficiently lipophilic to bioaccumulate in organisms, they are also subject to biotransformation processes and so do not appear to biomagnify through food chains [18], including those in the Arctic [13].

1.2. Sources, production, use and trends

Anthropogenic sources of combustion related PAHs within the Arctic are isolated and although they may be important on a local scale [19] they are generally negligible compared to the deposition of PAHs originating from the burning of fossil fuels and biomass at lower latitudes [3,4,20]. At the high latitudes and low temperatures of the Arctic, deposition processes become more significant than evaporation [21] and atmospheric PAHs may undergo net deposition to surface water, soils and sediments. However, semi-volatile PAHs may also re-volatize from ground surfaces, resulting in secondary emissions of some PAHs to the Arctic atmosphere [9].

Given the widespread and ongoing emission of PAHs globally, the source(s) of Arctic contamination are difficult to derive empirically. Therefore, modeling approaches have been used to determine the predominant regions, activities and global transport processes influencing PAH concentrations in the Arctic (see Section 2.2). Additionally, because of the deposition of PAHs to surface media, sediment cores and ice cores have been used to investigate historical sources and trends (see Sections 3 and 4).

Pyrogenic PAHs are by-products of combustion and are predominantly emitted to the atmosphere via burning of fossil fuels (coal, petroleum, wood) and biomass (forest, grassland, or agricultural) [1]. Quantifying emissions is therefore one way to estimate the extent and source of environmental PAH releases.

The most recently published global atmospheric emissions inventories for PAHs are for 2004 and 2007 [22,23]. Both studies estimated global emissions based on the 16 USEPA priority PAHs. The estimated global emission of PAHs for 2004 was 520,000 tonnes (t) [22]. In 2007, the annual global atmospheric emission was lower at 504,000 t, the primary sources being biomass fuels, including firewood and crop residues (60.5%), open-field biomass burning (agricultural waste burning, deforestation, and wildfires, 13.6%), and petroleum consumption by on-road motor vehicles (12.8%) [23]. Asian countries contributed half (53.3%) of the global PAH emissions, with the largest emissions in 2007 coming from China (106,000 t) and India (67,000 t), followed by Brazil, Indonesia, Nigeria, Ethiopia, Pakistan, Congo Democratic Republic, Vietnam, and Russia [23].

Emissions of the 16 USEPA priority PAHs in circumpolar and neighboring countries in 2007 based on estimates from Shen et al. [23] are shown in Fig. 2. These countries (including a category showing the combined northern European countries except for Norway, Sweden, Denmark and Finland) emitted 150,000 t of PAHs in 2007 or about 30% of global emissions.

Source profile estimates for Russia and the USA, the two largest PAH emitters among the circumpolar countries are shown in Fig. 3. Motor vehicles and wildfire sources were important in both countries although indoor firewood burning was estimated to be much more important in the USA than in Russia.

Shen et al. [23] also calculated historical global PAH emissions from 1960 to 2008 and projected future emissions through 2030. According to their analysis, total PAH emissions peaked globally in 1995 at 592,000 t and then declined reaching 499,000 t in 2008. Future global emissions are projected to continue declining, by 46–71% by 2030.

2. Environmental fate

2.1. Transformation processes

PAHs are not always present in the environment as parent compounds; hydroxylated-, nitro- and oxy-PAH derivatives may form during combustion or from abiotic and biotic transformation processes [9,24,25]. Microbial activity in soil, sediments and sea ice alters PAH profiles in the abiotic Arctic environment. Dong et al. [26] reported PAH concentrations in Arctic Ocean sediment samples collected from four sites on the Chukchi Plateau to the Makarov Basin in summer 2010. Concentrations of the 16 USEPA priority PAHs varied from 2.0 to 41.6 ng/g dw, and decreased with sediment depth and movement from the southern to the northern sites. Dong et al. [26] also identified several bacteria involved in PAH degradation *in situ*. The potential degraders included *Cycloclasticus*, *Pseudomonas*, *Halomonas*, *Pseudoalteromonas*, *Marinomonas, Bacillus, Dietzia, Colwellia, Acinetobacter, Alcanivorax, Salinisphaera and Shewanella*, with *Dietzia* the most abundant and present in all sediment samples. *Cycloclasticus* and *Pseudomonas* showed the best PAH degradation capability under low temperatures. They concluded that PAHs and PAH-degrading bacteria were widespread in the deep-sea sediments of the Arctic Ocean. Bagi et al. [27] reported on the biodegradation of the PAH naphthalene, which in addition to being a combustion byproduct is found in abundance in crude oil, and determined the biodegradation rate, temperature response and bacterial community composition of seawaters from two climatically different areas (North Sea and Arctic Ocean). Three-fold higher naphthalene degradation rates were observed in Arctic seawater samples as compared to those from the North Sea, suggesting that the biodegradation capacity in cold seawater is not necessarily lower than that of temperate seawater, as is often assumed. Garneau et al. [28] found that Arctic sea-ice and sub-ice microbial communities are also capable of degrading PAHs. Ice-associated microbes collected from the Resolute Passage in the Canadian Arctic archipelago were incubated with crude oil in a controlled microcosm experiment. Levels of 2-, 3- and 4-ring PAHs and their methylated analogs were measured five time points over the course of the 15-day experiment. Both sea-ice and sub-ice microbial communities produced a constant loss of

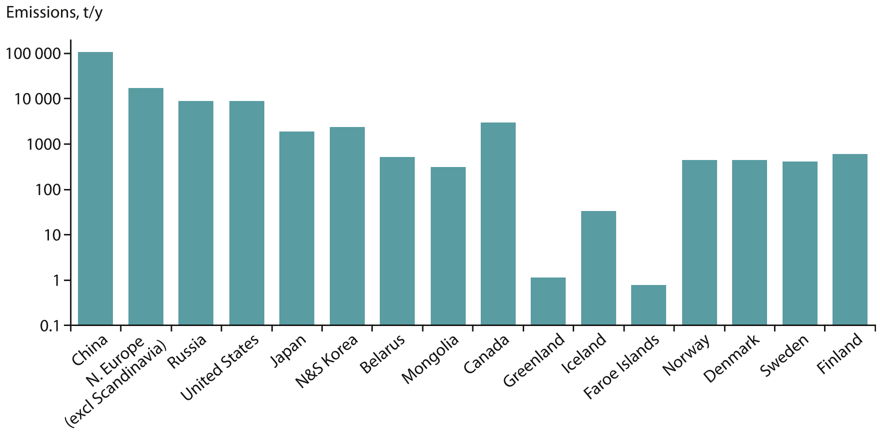

Fig. 2. Emissions of the 16 USEPA priority PAHs in circumpolar and neighboring countries in 2007 based on estimates from Shen et al. [23]. Estimated values for Scandinavian countries (Greenland, Iceland, Faroe Islands, Norway, Denmark, Sweden and Finland) are provided separately from those of northern Europe. Source: AMAP [14].

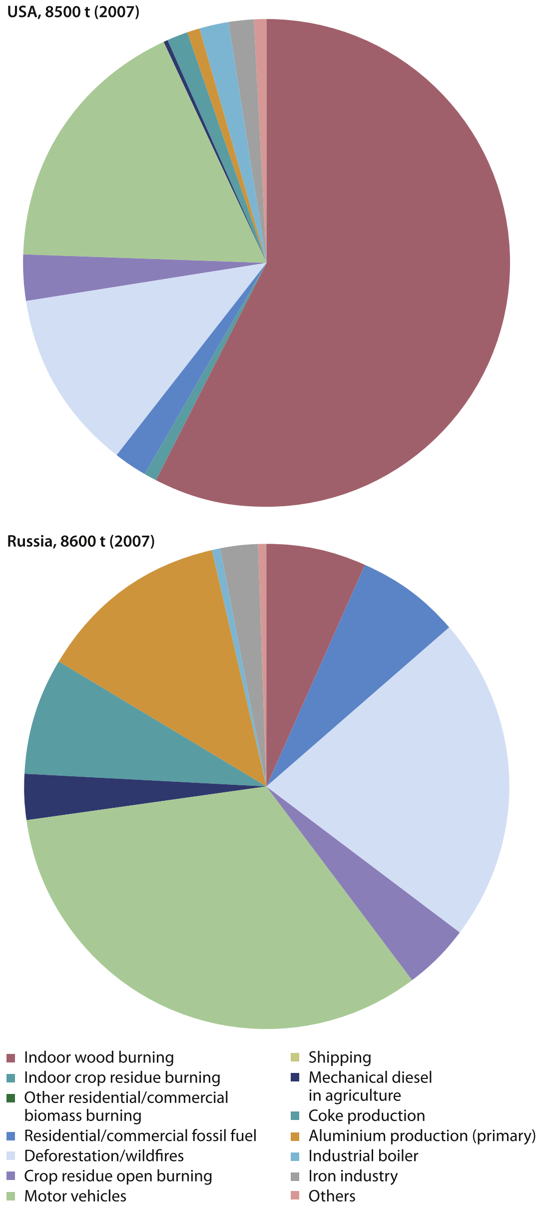

Fig. 3. Emission source profiles of the 16 USEPA priority PAHs in the USA and Russia. Sources: Shen et al. [23]; AMAP [14].

parent and methylated PAHs throughout the incubation period; however, sub-ice seawater microorganisms produced a larger overall decrease of PAHs (~70–80% loss) compared to sea ice-associated microorganisms (~30–40% loss) by the end of the experiment.

PAHs are actively biotransformed by vertebrates and many invertebrates via the cytochrome P450 enzyme system. However, several recent studies indicate that PAHs may bioaccumulate in some Arctic invertebrates due to a reduced ability to metabolize and eliminate these compounds. Experimental exposures of the high-Arctic copepod (*Calanus hyperboreus*) to PAHs including pyrene, phenanthrene, and benzo[a]pyrene (BaP) identified only trace levels of the primary metabolite, 1-hydroxypyrene, suggesting these lower-trophic organisms have a limited capacity to eliminate these PAHs, leading to their overall bioaccumulation [29–31]. Experimental exposures of the marine amphipod, *Gammarus setosus* to pyrene identified two metabolites – the glucose and sulfate conjugate of 1-hydroxypyrene, however, there was no appreciable decrease of the total metabolites or body burden of pyrene over the course of the 96 h depuration period, suggesting a potential of the organism to biotransform, but not completely eliminate pyrene [32].

Many vertebrates, including fish and marine mammals, are capable of efficiently metabolizing PAHs, thus tissue levels of parent compounds are typically low [33]. Quantifying PAH metabolites is therefore a generally accepted method for estimating biotic exposure [34]. Specifically, CYP1A1-mediated phase I biotransformation results in hydroxylated PAH metabolites (OH-PAHs) which are considered adequate markers of exposure to parent PAHs, and are also capable of causing adverse effects. Hydroxylated PAHs have been measured in several Arctic fish species (see Section 3.4).

Derivatives of parent PAHs (nitro-, oxy- and OH-PAHs, as well as many other oxygenated species such as aldehydes and quinones) may be formed during combustion and released as primary emissions, or may be secondarily formed via photochemical reactions in the atmosphere. The topic has been thoroughly reviewed by Keyte et al. [9] and Atkinson and Arey [35]. Parent PAH atmospheric residence times are generally hours or days at most, due to reaction with the hydroxyl radical in the gas-phase. OH–PAHs are formed through hydroxyl radical addition to the PAH. Nitro-PAHs can be produced by reaction of nitrate radical as well as from the reaction of OH-PAHs with nitrogen dioxide and loss of a water molecule. Nitro-PAHs can also be formed via HNO_3-catalyzed NO_2 reaction in the particulate phase. Oxy-PAHs may are formed through initial OH radical reactions in the gas-phase or ozone reaction in the particulate phase. For the relatively volatile two- and three-ring PAHs, reactions in the gas phase are likely to dominate, while for compounds with four or more aromatic rings, reactions in both phases are important [11]. Nitro- and OH-PAHs may account for up to 20% and 10%, respectively, of the 16 USEPA PAH concentration in air, and oxy-PAHs account for even more [11]. Nitro-PAHs have been mainly studied at urban sites, but have been detected at concentrations near detection limits ($< 0.01\ \mathrm{ng/m^3}$) at remote alpine locations [36]. The levels and spatial extent of nitro-PAHs in the Arctic atmosphere are unknown.

Photodegradation of PAHs also occurs in water by direct photolysis [37] or indirect photosensitization processes with transient excited species such as singlet oxygen ($^1O_2$), hydroxyl radical ($\cdot OH$), and other reactive species formed in sunlit natural waters [38]. Dissolved organic carbon can accelerate the photodegradation of small PAHs such as phenanthrene by enhancing the formation of reactive intermediates and inhibiting the photodegradation of large PAHs such as BaP by binding the PAH molecules [39]. OH-PAHs are one of the main photochemical transformation products of PAHs in water [40,41] and ice [42,43].

2.2. Modeling studies

The spatial/temporal distribution and long-range transport of atmospheric PAHs has been studied using measurements and models. Lammel et al. [16] used a multi-compartment chemistry-atmospheric transport model to investigate the influence of chemical transformation and gas/particle partitioning on the global distribution of PAHs in the atmosphere. The model is based on a general circulation model (GCM) with simplified atmospheric chemistry and an aerosol module that accounts for the aerosol components sulfate, black carbon, organic carbon, mineral dust and sea salt. Three PAHs (anthracene, fluoranthene, BaP) were used in model simulations and tested with several parameterizations of gas/particle partitioning and degradation assumptions. Model results indicated that in all scenarios, the mass fraction of the total environmental PAH burden in air is <4%; most of the global burden is stored in soils and on vegetation surfaces. Globally, PAHs are mostly distributed in and around source regions but do reach the Arctic through long-range atmospheric transport. The fraction of total environmental burden stored in the Arctic varied by scenario, but ranged from 0.5% to 12.8% for the three compounds tested. Overall, the simulations demonstrated that gas-particle partitioning in air has a substantial effect on the transport and environmental fate of PAHs, with a scenario that assumes adsorption onto organic matter and black carbon (soot) agreeing best with observations in Arctic regions.

To improve on model predictions of atmospheric transport of PAHs to remote regions such as the Arctic, Mu et al. [44] tested an advanced kinetic model framework (ROI-T scheme) that accounts for the influence of temperature and humidity on the phase state, diffusivity, and reactivity of multiphase PAHs by gaseous ozone. The transport of BaP from East Asian source regions to the Arctic was modeled using the new ROI-T scheme and other commonly used schemes (e.g., Kwamena scheme) and results were compared with observational data to evaluate model performance. The ROI-T model produced better predictions at all sites, including source regions and mid-latitude regions, but most significantly at Arctic sites, where air masses underwent the longest and coldest transport. In agreement with observational data, the model also estimated larger fluxes of BaP to the Arctic than previous models, likely by accounting for the lower degradation rates experienced at the low temperatures of higher atmospheric altitudes. This ‘freezing effect’ produces a longer lifetime of BaP in the atmosphere (20 days) as compared to estimates of earlier models (2–3 h), suggesting that the global transport of BaP from source regions may be more efficient that previously thought.

Wang et al. [3] used a probabilistic model based on backward air mass trajectory calculations to track the sources and atmospheric pathways of PAHs to the Canadian High Arctic. An integrated source contribution function (ISCF) that included air mass movements, emission intensities at sources, and the processes of partitioning, indirect photolysis, and deposition, was used to predict the contributions of various source regions for a total of 15 PAHs. Model results were further validated using air concentrations measured at the Alert air monitoring station in northern Ellesmere Island, Nunavut, throughout 2004. According to the model results, almost all PAHs detected in air at Alert originated from the northern hemisphere, with the three major source areas of eastern Russia/Asia, northern Europe/western Russia, and North America accounting for 25%, 45%, and 27% of atmospheric concentrations, respectively. Northeast China accounted for only 2% of the total. They concluded that the most important emission activities that resulted in PAH transport to the Canadian High Arctic were biofuel combustion (24%), followed by aluminum electrolysis (22%), and domestic coal burning (21%).

PAH concentrations and source regions also vary seasonally. In the summer, PAH concentrations are orders of magnitude lower than in the winter and spring when the model predicted more frequent long-range atmospheric transport events. According to the model, the majority of atmospheric PAHs in the summer and autumn are from northern Canada and North American sources, respectively; however in winter and spring, Russia and Europe are the major sources, with PAHs arriving in conjunction with the well-known Arctic haze events.

Friedman and Selin [4] gave additional evidence for the seasonality of PAH transport to the Arctic using a chemical transport model (GEOS-Chem) that includes parameters for uncertain PAH properties, such as oxidation, gas-particle partitioning and deposition. Using a model driven by assimilated meteorology from the NASA Goddard Earth Observing System (GEOS), the global transport of three representative PAHs (phenanthrene, pyrene and BaP) was simulated for the period 2004 to 2009. Model results showed PAH concentrations over both non-urban mid-latitude sites and the Arctic were significantly higher in winter than in summer. However, the Arctic experiences stronger seasonal variations, possibly reflecting increased seasonal variation in oxidation or transport, or the effect of springtime Arctic haze. Simulations based in Spitsbergen, Norway were also used to attribute atmospheric PAH concentrations in the Arctic to different source regions. Model results showed European emissions contributed the most (47–70% for the three compounds investigated), followed by emissions from Russia (13–29%) and North America (9–15%). East and South Asian emissions combined contributed just 1–8%. Overall, European and Russian emissions combined accounted for more than 80% of episodic high-concentration events at Spitsbergen.

The same GEOS-Chem model was used to evaluate the effects of future changes in climate and emissions (separately and together) on atmospheric PAH concentrations in the Arctic [45]. Model simulations compared the global transport of the same three representative PAHs (phenanthrene, pyrene and BaP) under present-day climate and emissions, and those projected for 2050. Model results showed future changes in simulated atmospheric PAH concentrations will be driven by declining anthropogenic emissions, with declining concentrations projected for each PAH simulated. However, the declines projected are greater in the northern hemisphere mid-latitudes (up to 38%) than for the Arctic (up to 8%). Differences in PAH volatility will influence how individual compounds respond to future climate scenarios, as behavior of gaseous PAHs is controlled primarily by competition between increasing deposition and increasing re-emission. Concentrations of volatile PAHs may increase in response to climate change because re-emission increases are projected to outweigh deposition increases, while the opposite is the case for particle-bound PAHs. Overall, the model results suggest the High Arctic is a priority area for resolving the influence of a changing climate versus anthropogenic activities on atmospheric PAHs and emphasize the importance of improving long-term measurements in this region.

To estimate the flux of PAHs between air and seawater, Ke et al. [46] developed a level III fugacity model using observational measurements collected from the Bering Sea in summer. The transfer fluxes across the air-sea interface estimated for seven PAHs (acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene and pyrene) were always higher for air-to-seawater than those from seawater to air, suggesting a net deposition of PAHs to seawater, and that the oceans serve as a sink for PAHs, at least in summer. However, deposition and volatilization of two PAHs, acenaphthylene and acenaphthene, were close to equilibrium. Since temperature plays a significant role in the air-water exchange processes of POPs, the environmental changes expected to accompany global climate change have the potential to disrupt this equilibrium, possibly leading to remobilization of these compounds from reservoirs in the Arctic Ocean.

3. Environmental concentrations

3.1. Air and precipitation

3.1.1. Air and aerosol

PAHs have been monitored in air at the AMAP stations of Alert (82°30′N, 62°20′W), Zeppelin (78°54′N, 11°53′E) and Pallas (68°00′N, 24°15′E) since 1992, 1994, and 1996, respectively [47]. Fig. 4 shows the range in air concentration for four PAHs (phenanthrene, pyrene, BaP and indeno[1,2,3-*c*,*d*]pyrene) measured at each station. The median concentrations measured at Pallas were

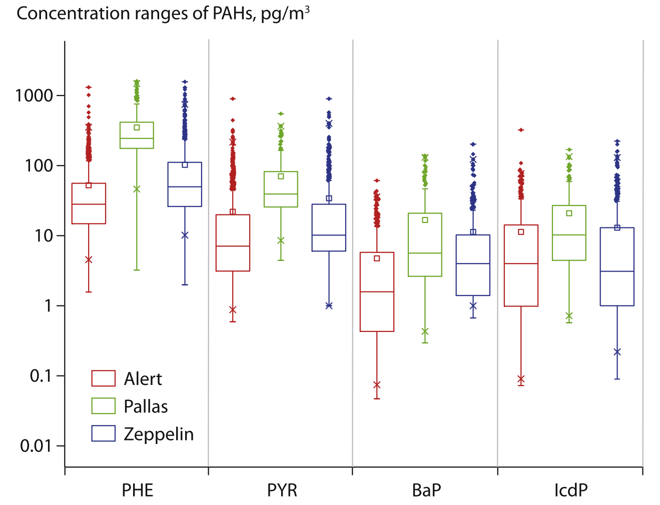

Fig. 4. Box-and-whisker plots of phenanthrene (PHE), pyrene (PYR), benzo[*a*]pyrene (BaP) and indeno[1,2,3-*c,d*]pyrene (IcdP) measured in air at Alert (1992–2012), Pallas (1996–2013) and Zeppelin (1994–2014). The boxes represent the 25th and 75th percentiles. The lines in the boxes and square symbols represent the median and the mean, respectively. All the outliers beyond the whiskers are shown individually. Sources: Yu et al. [47]; AMAP [14].

generally higher than those measured at the other two sites, which is probably due to its more southerly location and proximity to human activities in northern Europe.

At Zeppelin, the mean annual total PAH concentration (sum of 16 USEPA priority PAHs, Σ_{16}PAH) ranged from 500 pg/m^3 (2012) to 3530 pg/m^3 (1994) [48]. In 2013, the mean Σ_{16}PAH concentration was 1250 pg/m^3, the highest concentration observed since 2001 [48]. In 2014 and 2015 the mean PAH concentration was even higher, at 1430 pg/m^3 and 1300 pg/m^3, respectively [49,50]. At Alert, Σ_{17}PAH ranged from 3 pg/m^3 (1996) to 3000 pg/m^3 (1992) [51]. At Pallas, Σ_{12}PAH ranged from 54 pg/m^3 (1998) to 3900 pg/m^3 (2006) [51].

Fig. 5 summarizes the time series for phenanthrene and BaP concentrations measured at Alert, Pallas and Zeppelin [47]. At Alert, concentrations of both compounds declined between 1992 and 1999 (statistically significant for phenanthrene, $p < 0.05$; but not for BaP, $p = 0.078$) and then rose again to peak in 2004. This was followed by a statistically significant decline ($p < 0.05$) up to the end of 2012. A similar pattern was observed for phenanthrene at Pallas but the decline was not significant ($p > 0.05$). BaP appeared to decline in the early 2000s (statistically non-significant, $p > 0.05$) but concentrations have since increased and became more variable after 2010. At Zeppelin, a continuous and statistically significant ($p < 0.05$) decline in concentration was observed for phenanthrene, but no change was observed for BaP in the last two decades.

Air samples collected from Alert between February and June 1991 were used to examine the influence of the polar sunrise on atmospheric PAH levels [52]. Total suspended particles were collected weekly using a high volume air sample prior to, during, and following the polar sunrise that occurs between March and April. Nineteen PAHs were identified in Arctic aerosol samples, with the highest concentrations detected in late February, before the polar sunrise (mean $\sum_{19}$PAH $= 850$ pg/m^3). Elevated PAH concentrations in winter are likely related to the long-range transport of polluted air masses from lower latitudes, the temperature inversion of the boundary layer that prevents pollutant dispersion, the presence of sea ice that inhibits the direct deposition of pollutants into the ocean, as well as lower aerosol elimination rates due to the absence of solar radiation.

Direct deposition of pollutants into the ocean, as well as lower aerosol elimination rates due to the absence of solar radiation. PAH concentrations generally decreased following the polar sunrise (mean $\sum_{19}$PAH $= 31.7$ pg/m^3). The dominant PAHs, fluoranthene and benzo[k]fluoranthene were ~36 and ~18 times less abundant after the polar sunrise. BaP and benzo[e]pyrene (B[e]P) were ~53 and ~43 time lower than before the sunrise; the ratio of BaP to BeP also declined, which is suggestive of photochemical degradation. Furthermore, concentrations of aromatic acids were correspondingly higher following the polar sunrise, potentially indicating the photochemical oxidation of PAHs as their source.

Several studies have reported PAH concentrations in Arctic air collected during research cruises departing from the North Pacific. Ding et al. [53] reported concentrations of 15 PAHs in gas and particle phase samples collected during a 2003 expedition from Bohai Sea (37.78°N, 123.12°E) to the Arctic (80.22°N, 146.75°W) between July and September 2003. The $\sum_{15}$PAH in air from Arctic sites were 928–6310 pg/m^3 in the gas phase and 23.2–2640 pg/m^3 in the particle phase. A decreasing latitudinal trend was observed for gas-phase PAHs, but not for particulate-phase PAHs. Ding et al. [53], concluded based on source attribution ratios for 16 priority PAHs, that coal burning was likely the predominant source of PAHs observed over the Arctic region in summer. However, it is quite possible that other combustion sources including forest fires could also be important (Ma et al., 2013).

Wang et al. [54] measured gas- and particle-phase PAHs in air from the North Pacific (38°30′N) to the Arctic Ocean (87°11′N) during July through September 2012. $\sum_{9}$PAH measured from the North Pacific to the Arctic Ocean showed little variability; the lowest concentration was recorded near Greenland (41.3 pg/m^3) and the highest near Hokkaido, Japan (112 pg/m^3). On average, gas-phase PAHs accounted for 79.5% of the total atmospheric PAH concentrations and significant linear correlations were observed between the gaseous concentrations of nine PAHs and latitude ($r^2 = 0.69$), indicating that the concentrations of gas-phase PAHs decreased with increasing latitude.

Ma et al. [55] measured 18 PAHs in marine boundary layer air collected during an Arctic expedition from the East China Sea to the High Arctic between June and September 2010. Atmospheric concentrations were highest in East Asia (30°–48°N, 4000 pg/m^3), followed by the Arctic Ocean (>70°N, 3400 pg/m^3) and the North Pacific Ocean (50°–66°N, 2400 pg/m^3). Source attribution ratios showed atmospheric PAHs originated from the combustion of biomass or coal. Proximity to primary sources, such as continental regions like East Asia, where high PAH concentrations were observed, and the influence of seasonal and regional sources (i.e., forest fires) appear to be the factors broadly controlling atmospheric PAH concentrations in the Arctic.

Arctic maritime air samples were collected in August/September 2008 during a six-week expedition departing from Longyearbyen, Svalbard and heading northward through the marginal ice zone and pack ice zone before reaching 87°30′N, roughly 150 nautical miles south of the North Pole. Concentrations of the 15 PAHs measured were very low and in many cases below detection limits, ranging from 7 to 240 pg/m^3 for Σ_{15}PAH and 0.1–100 pg/m^3 for single components [56].

In 2011 and 2012, atmospheric PAHs were measured in the vicinity of two coal-fired power plants in Longyearbyen and Barentsburg (Svalbard), Norway [57]. The highest PAH concentrations in air were found near Longyearbyen power plant (62 μg/m^3 ΣPAH), with BaP and benzo[*a*]fluoranthene being the major contributors to the ΣPAHs. The PAH profile directly emitted by the plant was dominated by indeno[1,2,3-*c,d*]pyrene, benzo[*g,h,i*]fluoranthene, benzo[*b,j,k*]fluoranthene, benzo[*a*]fluoranthene, BeP and BaP. However, air samples collected at different distances from the

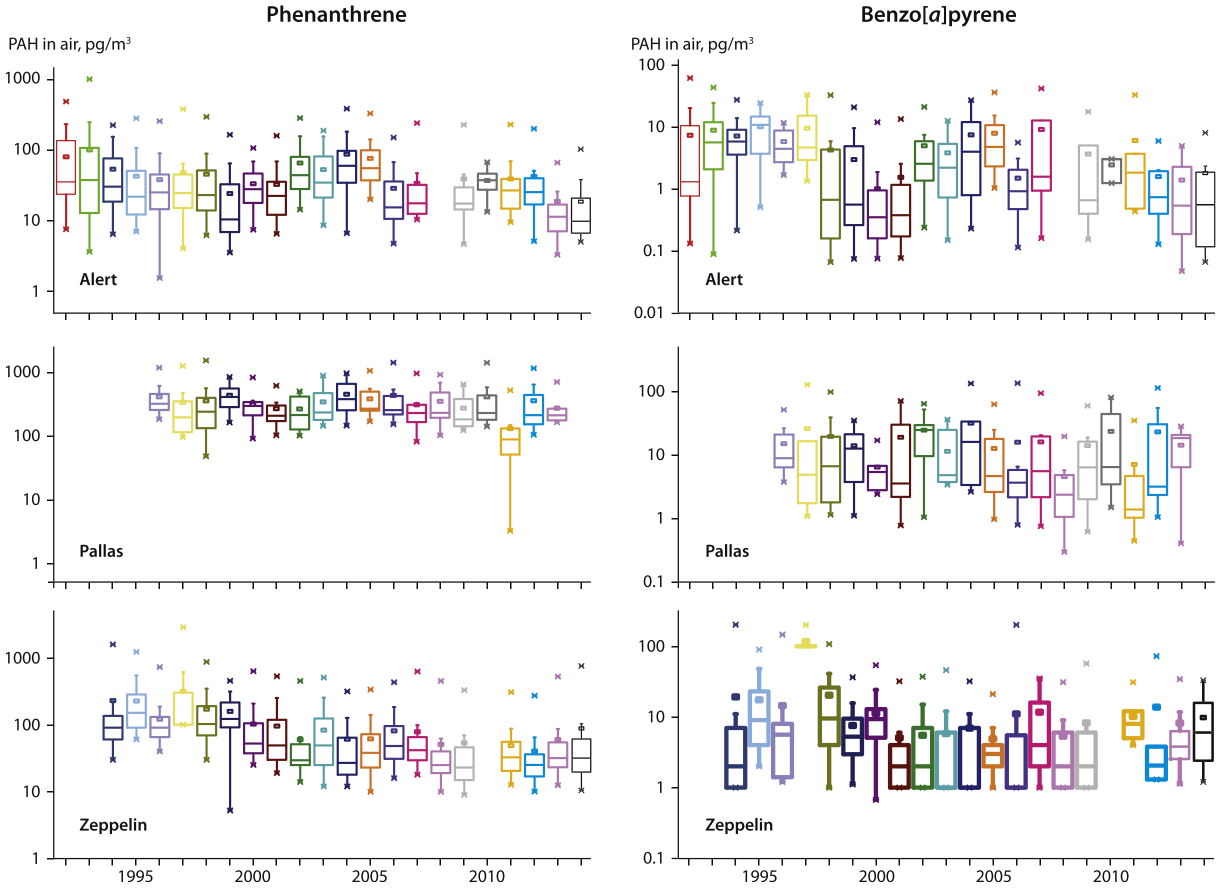

Fig. 5. Time series of phenanthrene and benzo[a]pyrene (BaP) observed in air at Alert, Pallas and Zeppelin. The boxes represent the 25th and 75th percentiles annually. The lines in the boxes and square symbols represent the annual median and annual mean, respectively. All outliers beyond the whiskers are shown individually. Sources: Yu et al. [47]; AMAP [14].

power plant appeared to only partially reflect this PAH profile and varied widely between sites [57].

Passive air sampling of PAHs in Alaska using XAD resin samplers was conducted as part of the Western Airborne Contaminants Assessment Project (WACAP) [58,59]. Samplers were deployed for 1 year (2005–2006) at five locations within national parks in Alaska and the western US lower 48 states. Seventeen PAHs (the 16 USEPA priority PAHs except for naphthalene and including BeP) and retene, an indicator for biomass burning, were determined. Almost all PAHs were below method detection limits (<MDL) (approximately <10 pg or 0.2 pg/g in dry XAD) at all sites. Phenanthrene and pyrene were elevated at the Noatak National Preserve on the Alaska North Slope (Tables S–1). Overall, PAH concentrations were lower than in samples from national parks in the western US States but difficult to compare with high volume air sampling because they were not reported on an air volume basis. The samplers were also designed to sample gaseous PAHs and at prevailing temperatures most of the 3- to 5-ring PAHs would have been particulate bound.

3.1.2. Snow

A suite of organic compounds was measured in snow from the Greenland Ice Sheet at Summit, Greenland (72°N, 38°W, 3200 m elevation) in summer 2005. A 3-m snow pit was sampled at 20-cm increments to generate a profile of PAHs spanning from 2002 to 2005. Three PAHs known to originate from combustion sources (phenanthrene, fluoranthene, and retene) were detected at low concentrations and total PAHs were highest in the surface layer (1.9 ng/kg), representative of 2005 [60].

As part of WACAP, PAHs were analyzed in snow from Denali National Park and Preserve (Denali NP&P), Gates of the Arctic National Park (GAAR), and Noatak National Preserve (NOAT) in Alaska, as well as in snow from more southerly Alaskan and western US national parks from 2003 to 2005 [58,61] (Fig. 6; Tables S–2). Total PAH concentrations were low, ranging from 0.3 to 17 ng/L at all six locations in Alaska north of 60°N. Fluoranthene and pyrene predominated in snow from the Denali NP&P catchments while benzo[*b*]fluoranthene and phenanthrene predominated in snow at the more northerly NOAT and GAAR locations. Annual fluxes of ΣPAH_{16} at Wonder Lake catchment (0.005 $\mu g/m^2$) and McLeod Lake (0.03 $\mu g/m^2$) in the Denali NP&P in 2003 were more than 10-fold lower than in western US parks and the Lake Matcharak catchment in GAAR (0.1 $\mu g/m^2$). However, snow in Burial Lake catchment (Noatak NP), the most northwesterly site, had 40-fold higher ΣPAH_{16} fluxes than at Lake Matcharak and were maybe affected to a greater extent by long-range atmospheric transport from Asia in winter [61].

Atmospheric concentrations may also be influenced by the temperature-dependent deposition and volatilization from snow or soil to air. The seasonality of air-soil/snow exchange of PAHs was examined through weekly measurements at an urban arctic site in Tromsø, Norway over fourteen months between May 2013 and July 2014 using a soil fugacity sampler [62]. Total concentrations of 53 PAHs in ranged from 64 to 460 pg/m^3 in air (at 1.5 m height), and 67–290 ng/g dw in surface soils. A comparison of the fugacity of PAHs in air (f_a) and snow/soil (f_s) allows the direction of soil/snow air exchanges (deposition vs volatilization) to be determined. The

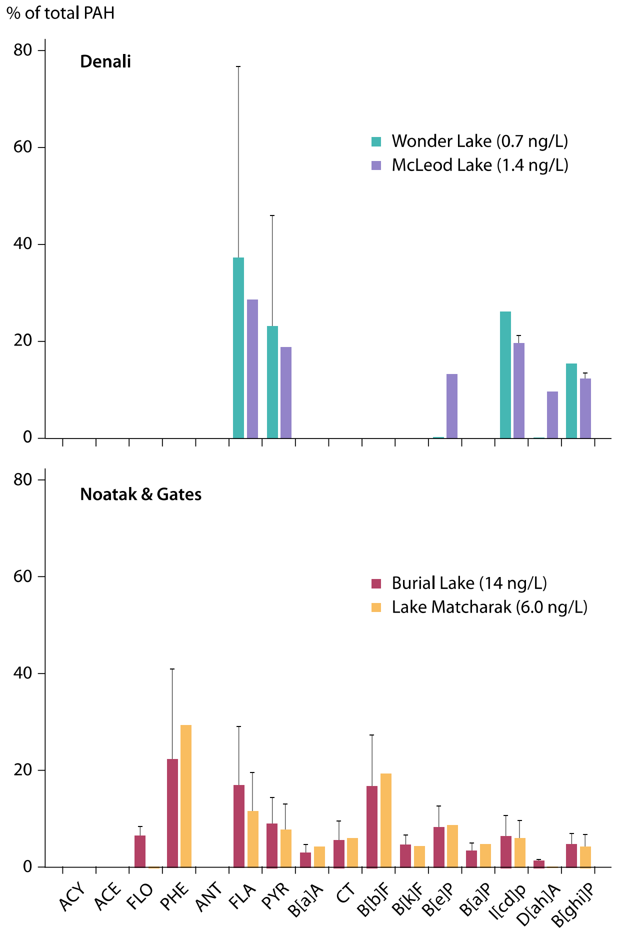

Fig. 6. PAH profiles measured in the seasonal snowpack from four lake catchments in Alaska national parks. Bars represent the average percentage of total PAH concentration and error bars are relative standard deviation. Total PAH concentration for each lake catchment is provided in parentheses. Abbreviations: ACE: acenaphthene, ACY: acenaphthylene, ANT: anthracene, B[a]A: benzo(a)anthracene, B[a]P: benzo(a)pyrene, B[b]F: benzo(b)fluoranthene, B[e]P: benzo(e)pyrene, B[ghi]P: benzo(ghi)perylene, B[k]F: benzo(k)fluoranthene, CT: chrysene+triphenylene, D[ah]A: dibenz(a,h)anthracene, FLA: fluoranthene, FLO: fluorene, I[123-cd]p: indeno(1,2,3-cd)pyrene, PHE: phenanthrene, PYR: pyrene. Sources: (Usenko et al. [61]; AMAP [14].

average fugacity ratios (ln f_s/f_a) for PAHs were near equilibrium (between -1.2 and 0.53) from January to March, and higher than 0.53, indicating volatilization during the rest of the year, with the highest fugacity ratios detected in June and July of 2013 and 2014. The low ratios observed in winter reflect the high sorbing capacity of snow for PAHs, while the higher fugacity ratios observed in warmer months support the role of soils as a source of PAHs contributing to atmospheric concentrations.

3.2. *Terrestrial environment*

Several studies have investigated levels of PAHs in terrestrial soils at Arctic sites with known anthropogenic impacts in comparison to nearby, unimpacted sites. Abakumov et al. [63] compared levels of 15 PAHs in soils from former and present research stations in the Russian Arctic to those in soils collected from unimpacted sites located several kilometers away. At most paired sites, PAH levels were higher near research stations compared to closely located natural sites. Marquez et al. [64] compared levels of the 16 USEPA PAHs in soils collected around an abandoned mining town, Pyramiden, Svalbard (Norway). With the exception of two unimpacted reference sites, several kilometers away, soil samples were collected near the center of town, in proximity to coal and diesel power plants, a heliport, and harbor. ΣPAHs in soil at seven impacted sites ranged from 186 to 11600 ng/g dw (mean concentration 3170 ng/g dw) compared to 52 ng/g dw at the unimpacted sites. Attribution ratios pointed to a mix of pyrogenic and petrogenic PAH sources across the study area, including at the unimpacted reference site. The authors speculate that petrogenic sources could include both natural coal deposits and past coal extraction activities within the area. Collectively, these studies emphasize the potential contribution of local sources to PAH pollution within the Arctic as well as the challenge of distinguishing contributions from natural sources such as coal deposits.

Surface soil, moss, and reindeer dung were collected simultaneously in July and August 2007 from 12 sites at Ny-Ålesund, Svalbard and analyzed for 16 standard unsubstituted PAHs [65]. Σ_{16}PAH concentrations were 37–324 ng/g dw in soil, 158–244 ng/g dw in moss, and 49–340 ng/g dw in reindeer dung. Of the three compartments, soil was enriched with the more high molecular weight PAHs (4–6 rings), and had less of the low molecular weight PAHs (2–3 rings) than moss and reindeer dung. The difference in distribution between the three compartments is probably related to the physical-chemical properties of individual PAHs and their different accumulation routes; whereas soil accumulates PAHs mainly through dry/wet particle deposition, moss sequesters PAHs mainly from the gas phase. Interestingly, the PAH profiles of reindeer dung and moss, the main food source for reindeer, were not significantly different [65]. In addition, the overall proportions of 2- and 3-ring PAHs relative to ΣPAHs at Ny-Ålesund were higher than published values from non-Arctic sites, which is consistent with a long-range atmospheric transport origin of low molecular weight compounds. A long-term (1987–2006) study of reindeer from Abisko, Sweden reported that PAHs were below the detection limit in muscle [66].

As part of WACAP, PAHs were analyzed in conifer needles and lichen from samples from four Alaskan national parks north of 60°N, as well as at more southerly Alaskan and western US national parks [58,59]. Most of the 16 PAHs were at concentrations below MDLs (Tables S–3). Fluoranthene, fluorene and phenanthrene were detectable especially in samples from Katmai NP and Wrangel-St Elias NP, both in southern Alaska. Retene was detected at five of six sampling locations at average concentrations of 1.5–7.6 ng/g dw. Concentrations of retene were highest in national parks in forested areas of southern Alaska and lowest in GAAR which is north of the tree-line on the Alaskan North Slope (Tables S–3). Given that sampling occurred in August, the retene was probably from forest fires in the region.

Muscle samples from moose harvested in Denali NP&P were analyzed as part of WACAP (Tables S–4). Eight of the 16 PAHs were detectable at low ng/g ww concentrations. Acenaphthene was the predominant PAH averaging 9.3 ng/g ww (in four of six samples) while the second most common PAH was indeno[1,2,3-*cd*]pyrene at 1.25 ng/g ww (in two of six samples).

3.3. *Freshwater environment*

3.3.1. *Surface water*

Surface waters were sampled from 25 streams in the remote Fuglebekken basin in southern Spitsbergen, Svalbard during July 2009. Total concentrations of 12 PAHs were 4–600 ng/L, with the highest levels detected at the base of the mountain that seasonally receives snow and ice meltwater. Indicator ratios suggest that long-range transport of combustion-related PAHs was the predominant source of PAHs to the basin's surface waters [67].

A suite of PAHs (phenanthrene, anthracene, fluoranthene, fluorene, pyrene, benzo[*a*]anthracene, benzo[*b*]fluoranthene, benzo[*k*]fluoranthene, BaP, dibenz[*a,h*]anthracene, benzo[*ghi*]perylene and indeno[1,2,3-*cd*]pyrene) were investigated in surface water from Lake Abiskojaure in northern Sweden [68]. Four PAHs were detected: phenanthrene (370 pg/L), fluoranthene (99 pg/L), fluorene (180 pg/L) and benzo[*g,h,i*]perylene (23 pg/L). At Kalixälvens gruvsamhälle (an old mining village at the Kalixälv river in Lapland, point source) six PAHs were detected in surface water: phenanthrene (290 pg/L), fluoranthene (140 pg/L), fluorine (180 pg/L), pyrene (71 pg/L), benzo[*b*]fluoranthene (8 pg/L) and benzo[*g,h,i*]perylene (15 pg/L).

Törneman et al. [69] studied EU Water Framework Directive priority substances mostly in water samples from Lake Abiskojaure in northern Sweden. Eight PAHs (indeno[1,2,3-*cd*]pyrene, naphthalene, anthracene, BaP, benzo[*b*]fluoranthene, benzo[*ghi*]perylene, benzo[*k*]fluoranthene, fluoranthene) were investigated in water samples every month in 2008. Only one sample contained detectable concentrations of one PAH, naphthalene (12 ng/L).

As part of WACAP, PAHs were analyzed in lake water from four lakes within national parks north of 60°N as well as in as more southerly Alaskan and western US national parks. Samples were collected in 2004 using filter/modified SpeedDisk systems [58,70]. Despite good detection limits due to large volume samples (25–50 L), all 16 USEPA PAHs were below MDLs (~0.4–9 pg/L) in almost all samples. The exception was phenanthrene which was detected in McLeod Lake (86 pg/L) and Wonder Lake (2400 pg/L) in Denali NP&P. Those concentrations are comparable to those detected in Lake Abiskojaure in northern Sweden.

3.3.2. *Sediment*

Sediments cores from four lakes within national parks north of 60°N in Alaska were analyzed for the 16 USEPA PAHs and retene [58,61]. Most of the 16 USEPA PAHs were below the MDL (typically <0.5 ng/g dw). Fluoranthene, benzo[*b*]fluoranthene, and phenanthrene predominated in three of the four lakes (Tables S–5) similar to the pattern in snow from the same catchments. McLeod Lake in Denali NP had the lowest ΣPAH concentrations with only acenaphthene detected. Lake Matcharak surficial sediment had ~17-fold higher ΣPAH fluxes than Burial Lake, mainly due to 10-fold higher phenanthrene concentrations (Tables S–5). This trend is the opposite of the snowpack ΣPAH flux measurements and suggests that the snowpack ΣPAH fluxes may vary significantly from year-to-year at these Alaskan sites.

Surficial sediments were collected from six remote lakes from Ny-Ålesund, Svalbard in 2005 [71]. Total PAH concentrations in lake sediments varied 100-fold, ranging from 11 to 1100 ng/g dw. Although Jiao et al. [71] cautioned that the exceptionally high PAH concentrations observed in two of the six lakes could be due to proximity to a coal mining operation, PAH indicator ratios suggested the majority of the PAHs were derived from pyrogenic sources.

Eide et al. [72] measured 22 PAHs in freshwater (and marine) sediment cores from southwestern and northern Norway covering the periods 1950–2002 and 1800–2004. The Σ_{22}PAH concentrations in surface lake sediments ranged from 217 to 7045 ng/g dw and were generally three to four times higher than at the marine sites. Source attribution ratios indicated that pyrogenic sources prevailed at most locations, with the exception of Svalbard, which exhibited high concentrations and patterns reflecting the area's active coal mining and combustion activities. The elevated PAH concentrations observed in Svalbard relative to other Norwegian marine environment locations have also been described elsewhere, but are not discussed here given their likely petrogenic source [73–76].

PAH measurements were obtained from freshwater sediments collected from the seven largest Arctic rivers to assess the role of fluvial transport of black carbon to the Arctic [77]. The samples were collected from the river deltas of the western Siberian lowlands (Ob and Yenisey Rivers), central Siberian uplands (Lena River), East Siberian highlands (Indigirka and Kolyma Rivers) and North America (Yukon and Mackenzie Rivers) in 2004 and 2005. Total concentrations of 18 PAHs were lowest in the Ob River (23 ng/g ww) and highest in the Mackenzie River (454 ng/g ww). Source attribution ratios indicated PAHs from the two North American rivers are likely to have originated from a petrogenic source, whereas those detected in the Russian Arctic rivers are likely to have originated from vehicle emissions (Lena River), wood combustion (Indigirka and Kolyma Rivers) and the combustion of grass, wood and coal (Ob and Yenisey Rivers) [77].

3.3.3. *Biota*

The 16 USEPA PAHs and retene were analyzed in whole fish samples from four lakes in Alaska [58]. Six of the 16 USEPA PAHs were detectable at low ng/g ww concentrations with lake trout (*Salvelinus namaycush*) from Wonder Lake in Denali NP&P having highest detection frequencies (Tables S–6). Lake trout from Lake Matcharak had 6-fold lower concentrations of ΣPAHs than in Wonder Lake despite nearly 3-fold higher concentrations in sediment (Tables S–5).

3.4. *Marine environment*

3.4.1. *Seawater*

Surface seawater samples were collected during the Fourth Chinese National Arctic Research Expedition from the East China Sea to the High Arctic (33.23–84.5°N) between June and September 2010 and analyzed for 18 PAHs [55]. Generally, seawater PAH concentrations decreased with increasing latitude. Mean concentrations of ΣPAHs were 150 ± 270 pg/L in East Asia (35–48°N), 76 ± 46 pg/L in the North Pacific Ocean (50–68°N), and 37 ± 18 pg/L in the Arctic Ocean (>70°N). Source attribution ratios suggest that the PAHs in seawater originated from a mixture of petrogenic sources, biomass combustion, and coal and liquid fossil fuel sources [55]. PAHs were less frequently detected in seawater than in air. Seawater PAH source profiles showed some similarity with those in air, which may be indicative of air-sea gas exchange and deposition of certain PAHs.

Lohmann et al. [78] analyzed surface seawater samples from 22 stations during a cruise across the North Atlantic and European Arctic. The highest PAH concentrations were detected in the southernmost sample (off Europe) and the two northernmost samples (around 82–84 °N). Overall, highest concentrations were determined for phenanthrene (10–180 pg/L), fluoranthene (10–130 pg/L), and fluorene (16–66 pg/L). Concentrations of anthracene, pyrene, benz[*a*]anthracene, benzo[*b*]fluoranthene and indeno[*g,h,i*]perylene were mostly <1 pg/L. For all PAHs, source attribution ratios indicated a combustion-derived origin for the PAHs detected, however, no explanation was offered for the elevated levels detected around 82–84 °N. Marine boundary layer air was also collected for analysis, but samples were considered to be cross-contaminated by ship exhaust and concentrations were not reported [78].

Surface seawater samples were collected from the temperate Pacific Ocean (34.4°N) to the Arctic Ocean (72.0°N) during the 4th Chinese National Arctic Research Expedition between June and September 2010 [46]. At sampling stations above (70°N), total concentrations of seven dissolved PAHs (acenaphthylene, acenapthene, fluorene, phenanthrene, anthracene, fluoranthene and pyrene) in surface seawater ranged from 1.5 ng/L to 2.2 ng/L.

Ke et al. [46] also determined the vertical profile of seven dissolved PAHs in seawater for a single site near the Chukchi Sea in the summer of 2010. Samples were collected at various depths extending from 0 m–600 m below the surface. Concentrations ranged from 1.7 ng/L at 615 m, to 2.4 ng/L at 30 m, and generally showed a decreasing trend with depth, which is consistent with surface enrichment and deep depletion. The low salinity of the surface waters collected at the site potentially indicate the higher levels of PAHs at shallow depths may have originated from the input of meltwater from continental glaciers or sea ice, rather than oceanic currents. Additionally, PAHs appeared to follow the same vertical distribution profile as chlorophyll, suggesting that phytoplankton may contribute to the transport of PAHs to deeper ocean waters.

3.4.2. Marine sediment

Sediment PAH data published prior to 2004 were extensively reviewed in a previous AMAP report [15]. The majority of reports thereafter come from the Norwegian Arctic and Barents Sea regions [71,72,79–82]. Although the regions under study overlap, detailed comparisons are difficult given differences in the number of compounds measured.

Measurements made throughout the Barents Sea suggest that anthropogenic inputs of PAHs are likely to be limited. Analyses of surface sediments and sediment cores collected in 2003 and 2004 from the western Barents Sea averaged 400–500 ng PAH/g dw [79]. In 2006, sediment cores collected from the southwestern Barents Sea in 2006 exhibited similarly low PAH concentrations, averaging 200 ng/g dw [83]. In both studies, source attribution ratios indicated PAHs were of a natural, mostly petrogenic origin, probably from natural seeps of oil-related hydrocarbons in the area [79,83]. Another study of sediment cores from the western Barents Sea detected similarly low concentrations of 12 PAHs (range, 35 ± 18 to 132 ± 66 ng/g dw). Based on concentrations detected in the deepest layers of the sediment cores, corresponding to the period prior to 1850, natural background PAH concentrations have remained fairly constant throughout the western Barents Sea over time [82]. However, in contrast to previous studies, a predominance of compounds indicative of atmospheric releases from aluminum smelters, coal and wood burning were detected [82].

Anthropogenic inputs of PAHs are becoming increasingly evident in Norwegian coastal sediments. In a study of the southwestern Barents Sea, Boitsov et al. [83] noted that samples collected near Norwegian fjords had PAH attribution ratios indicative of a pyrogenic origin, suggestive that coastal areas may be experiencing low inputs from human activities. Eide et al. [72] measured 22 PAHs in coastal sediments from Svalbard, Troms and Finnmark counties, Norway, covering the periods 1950–2002 and 1800–2004. Overall, Σ_{22}PAH concentrations in marine sediments ranged from 82 to 3076 ng/g dw, with the highest concentrations detected in Svalbard. Source attribution ratios indicated the PAHs originated from multiple sources, but were predominantly combustion related. Similarly, Boitsov et al. [80] measured PAH concentrations ranging from 9.5 to 1799 ng/g dw in surface sediments from the northern Norwegian coast (above 68°N) from 1992 to 1996 and found indications of predominantly pyrogenic sources. However, coastal sediments from two sites along the west coast of Spitsbergen, Svalbard from 2005 exhibited source attribution ratios solely indicative of a petrogenic source, and given the low PAH concentrations detected (25–38 ng/g dw), were attributed to natural processes such as seabed seeps [71].

New analyses are beginning to provide insight into the extent of PAH deposition to marine sediments across the Arctic. Deep-sea sediments were collected at four sites across the Arctic Ocean, including the Chukchi Plateau, Canada Basin, Alpha Ridge, and Makarov Basin [26]. Total concentrations of the 16 USEPA priority PAHs ranged from 2.0 to 41.6 ng/g dw and generally decreased with sediment depth. Concentrations also tended to decrease with increasing latitude, from the southernmost site (Chukchi Plateau; 41.6 ng/g dw) to the northernmost site (Makarov Basin, 2.0 ng/g dw).

3.4.3. Marine invertebrates

PAH data for marine biota prior to 2004 were extensively reviewed in a previous AMAP assessment report [15]. In 2011, Jörundsdóttir et al. [84] conducted a survey of PAH concentrations in blue mussel (*Mytilus edulis*) from remote sites in Greenland, Iceland, the Faroe Islands, Norway and Sweden and included urban sites for comparison. Overall, total concentrations of the 16 USEPA priority PAHs ranged from 28 to 480 ng/g dw, with the highest concentration detected in Ísafjörður harbor, a polluted site in northwestern Iceland. However, remote sites in eastern Iceland and western Greenland also had inexplicably high concentrations; 370 and 280 ng/g dw, respectively. Source attribution ratios and the predominance of pyrene in a large proportion of the samples was strongly indicative of a combustion source and atmospheric input [84].

In 2010, blue mussels and Iceland scallops (*Chlamys islandica*) were sampled from the Barents Sea near northern Norway (70°N) [85]. Concentrations of the 16 USEPA priority PAHs ranged from <MDL to 22 ng/g ww in blue mussel, and from 5.7 to 17 ng/g ww in scallops. Corresponding concentrations of 2–3 ringed PAHs, including naphthalene, anthracene/phenanthrene, dibenzothiophene and their alkylated homologues were generally higher at <MDL to 99 ng/g ww in mussels and <MDL to 65 ng/g ww in scallops.

3.4.4. Fish

PAHs are rapidly metabolized in fish, so quantifying PAH metabolites is considered the conventional method for estimating exposure in these species [34]. Specifically, the CYP1A1 phase I OH-PAHs are considered excellent chemical markers for exposure to the parent PAHs.

OH-PAH metabolites were measured in 60 liver samples of Arctic cod (*Boreogadus saida*) collected from offshore regions of the Canadian Beaufort Sea in 2012 [86]. Four-ring OH-PAHs were detected in 90% of samples with a mean concentration of 1830 ± 159.2 ng/g ww, while mean concentrations of 5/6- ring OH-PAHs in liver were lower (932 ± 104.3 ng/g ww) and detected less frequently (75%). The 2/3-ring metabolites were not detected. Because fish were sampled prior to any major anthropogenic activity (such as offshore oil and gas development) in the region, exposure to the parent PAHs was hypothesized to have originated from natural sources.

Baseline concentrations of nine PAH metabolites were measured in Arctic cod from pristine waters surrounding Svalbard in September 2001 and August 2002 [87]. All nine metabolites were detectable in cod bile with total concentrations averaging 1900 ng/g bile and 1940 ng/g bile for the two sites, Hinlopen Strait and Isfjorden, respectively. Of the nine metabolites analyzed, C2-phenanthrene, C1–OH-naphthalene, C2–OH-naphthalene and C3–OH-naphthalene were present at the highest concentrations, exceeding concentrations of 1-OH-pyrene by an order of magnitude.

Jörundsdóttir et al. [84] also measured OH-PAH metabolites in Atlantic cod (*Gadus morhua*) caught north of Iceland and along the Norwegian coast. Of the two metabolites measured, 1-OH-pyrene and 3-OH–BaP, only 1-OH-pyrene was found above the limit of quantification, and only in samples from the Norwegian coast, ranging from 44 to 140 ng/mL bile. 1-OH-pyrene was detectable, but not quantifiable in Atlantic cod bile from Iceland. The higher concentrations observed in Norwegian cod are probably due to a known point source of pollution near where they were caught. In contrast, the Icelandic cod were caught in the open sea where exposure to point sources of PAHs is limited [84].

In 2010, Atlantic cod from the Barents Sea near northern Norway (70°N) was analyzed for four OH-PAH metabolites, of which only two (1-hydroxypyrene and 1-hydroxyphenanthrene) were present in bile. The metabolites 3-OH—BaP and 2-hydroxynaphthalene were not detected [85].

3.4.5. *Seabirds*

Eggs from common eiders (*Somateria mollisima*), European shag (*Phalacrocorax aristotelis aristotelis*) and herring gulls (*Larus argentatus*) were collected from two remote islands, Sklinna and Røst on the Norwegian coast in 2012 and analyzed for the 16 USEPA priority PAHs [88]. Of these, five (naphthalene, anthracene, fluoranthene, pyrene, chrysene) were detected at concentrations above the limit of detection. Seabird eggs from Røst, located north of the Arctic Circle, had ΣPAH concentrations ranging from non-detectable to 29.6 ng/g ww. PAHs were not detectable in eggs of the European shag. In common eider and herring gull eggs, geometric mean PAH concentrations were 0.11 and 7.63 ng/g ww, respectively. Similar to results reported for Arctic fish, it is likely that PAHs are efficiently biotransformed in birds via CYP1A1 enzyme mediation to OH-PAHs.

3.4.6. *Marine mammals*

To our knowledge, no PAH data for arctic marine mammals are available in the published literature. However, concentrations in apex predators are generally low given that PAH concentrations tend to decrease with trophic level owing to biotransformation processes [13,18].

4. Environmental trends

4.1. *Spatial trends*

Broad-scale spatial analyses for PAHs in the Arctic are scarce. Yunker et al. [89] reported the first comprehensive analysis of persistent hydrocarbon tracers in sediment throughout the Arctic Ocean. A large suite of hydrocarbons, including parent and alkyl-PAHs, were measured in suspended particulates, sediment cores and surface sediments across the region. Spatial patterns in PAH distribution throughout the Arctic Ocean were investigated using multivariate analyses. Biomarker patterns revealed that central Arctic Ocean basin sediments are compositionally distinct from those of the Beaufort Sea and Barents Sea, but similar to those of the Laptev Sea. Source attribution ratios indicative of a combustion origin were detected in PAHs from suspended particulates in the Chukchi and Lincoln Seas and the central Arctic Basin. However, overall, Arctic sediments were overwhelmingly dominated by natural PAH inputs such that anthropogenic PAHs originating from combustion sources and atmospheric transport were deemed insignificant [89].

Similar results were found by Harvey et al. [90] and Foster et al. [91] who analyzed surface sediments from the Chukchi Sea (Alaska) and Baffin Bay (eastern Canada), respectively. Sediments from both locations exhibited PAH profiles overwhelmingly dominated by alkyl-PAHs, indicating a predominance of petrogenic sources and also that long-range atmospheric transport may be less important than natural inputs to sediments in unexploited regions [90,91].

Ke et al. [46] examined the latitudinal distribution of dissolved PAHs in surface seawater from the North Pacific Ocean to the western Arctic Ocean as part of an effort to develop a fugacity model for PAHs in (see section 2.1). Surface seawater samples were collected during a research cruise extending from the Japan Sea to the Arctic between June and September 2010, with samples collected in both the Bering and Chukchi Seas. Total concentrations of seven PAHs (acenaphthylene, acenapthene, fluorene, phenanthrene, anthracene, fluoranthene and pyrene) generally showed a decreasing trend with increasing latitude from 34.4°N (2.7 ng/L) to 72.0°N (1.3 ng/L), which is in agreement with previously reported spatial trends in atmospheric PAHs from Japan to the Arctic [53], suggesting that surface ocean currents, along with atmospheric transport may serve as another important pathway bringing PAHs from lower latitudes to the Arctic.

4.2. *Temporal trends*

A long-term monitoring project for atmospheric PAHs and other POPs at the High Arctic air monitoring station Alert in Nunavut, Canada has been ongoing since 1992 [92]. Between 1992 and 2002, PAH gas-phase concentrations of the 16 USEPA priority PAHs ranged from 113 to 516 pg/m^3 and particle-phase concentrations ranged from 38 to 392 pg/m^3. Although strong seasonal fluctuations obscured the detection of reliable temporal trends, general decreases in PAH concentrations were noted between 1993 and 1996, which strongly agrees with declines measured in atmospheric samples from mid-latitudes during the same period [93]. In a later study, Becker et al. [94] were able to resolve long-term temporal trends in PAH concentration at Alert using dynamic harmonic regression, a statistical time-series tool that identifies and removes seasonal components in the data (Fig. 7). Applying this technique to the analysis of atmospheric PAH data collected at Alert

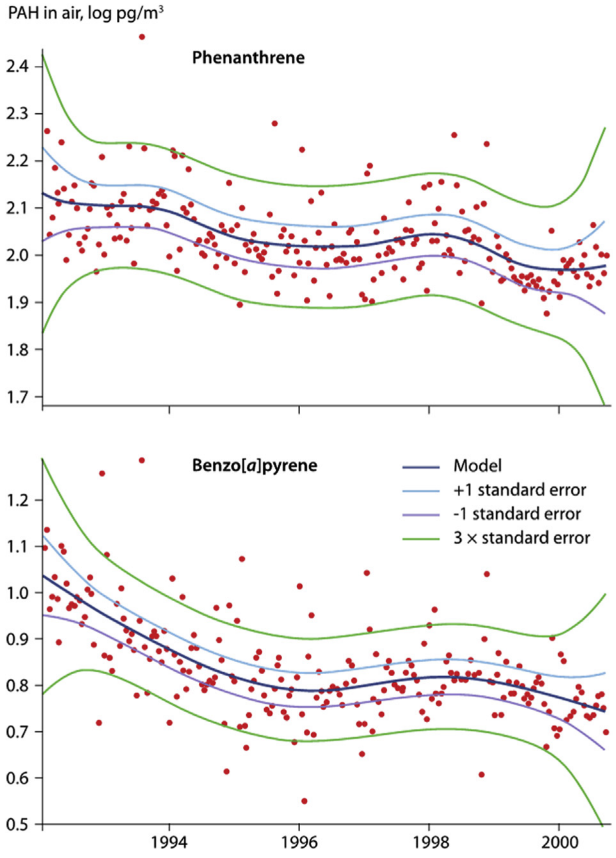

Fig. 7. Seasonally adjusted data and underlying long-term trends in two PAHs — phenanthrene and benzo[*a*]pyrene (BaP) — in air at Alert as identified by dynamic harmonic regression. Sources: Becker et al. [94]; AMAP [14].

between 1992 and 2000 identified seasonal patterns in PAH concentration with a consistent summer increase in many of the low-molecular weight (two-to three-ringed) PAHs. Furthermore, removing seasonal trends revealed a decline in PAH concentration over the period 1992–2000. While lighter PAHs exhibited linear declines throughout the study period, many of the higher molecular weight PAHs demonstrated early declines followed by a leveling off of concentration by the mid/late 1990s [94].

De Laender et al. [13] reconstructed temporal trends for PAHs and other legacy POP classes in the Barents Sea and Norwegian Sea using a food web bioaccumulation model to derive complete data sets. Monitoring data for 21 species occupying various trophic levels was extracted from the International Council for the Exploration of the Sea (ICES) database and used to infer temporal trends for POPs in seawater and biota between 1985 and 2010. However, temporal trends for PAHs were based on concentrations in mussels only, since trend data were not available for other species. The analysis indicated that concentrations of legacy POPs in the Barents/Norwegian Sea fauna decreased 10-fold over the past 25 years, while concentrations of fossil fuel-derived PAHs increased 10- to 30-fold in seawater and lower trophic level biota (mussels and fish). The trend analysis also indicated that in the last five years of the time range considered (2005–2010), PAH concentrations stabilized, potentially indicating a recent moderation in global fossil fuel emissions [13].

Eide et al. [72] measured 22 PAHs in freshwater and marine sediment cores from southwestern and northern Norway covering the periods 1950–2002 and 1800–2004. The PAH concentrations in lake sediments of northern Norway have increased in recent decades, although concentrations are still much lower than those from the south. In contrast, in southwestern Norway, ΣPAHs increased from 1800 to 1950, but then decreased by a factor of four. Eide et al. [72] attributed the increase in sediment PAH concentrations to the growth in global industrialization over this period. The decrease in sediment PAH from the 1950s onward coincides with the reduction in heath burning and reduced long-range transport of pollutants from Europe. In comparison to freshwater sediments, marine sediments exhibit less pronounced temporal trends, probably due to the greater influence of perturbation by ocean currents, ship traffic, and oil spills.

Similar trends were found by Boitsov et al. [83] who measured PAHs in sediment cores from the southwestern Barents Sea in 2006. An almost 10-fold increase in concentration was observed in upper sediment layers, representing the 1910–1940s period. Thereafter, concentrations level off and, from about the 1980s onward, decrease slightly in surface sediments.

Another study investigated PAH concentrations and profiles in Barents Sea sediments from the last 10 years [81]. Surface sediment samples were collected from the Barents Sea in the period 2001–2005 and compared with data previously obtained from sediments collected in 1992–1998. Five unique areas were identified within the Barents Sea with regard to their PAH concentrations and profiles, but for most of the regions no significant differences were found between the two periods studied. However, in the southeastern Barents Sea, pyrogenic PAH concentrations and source attribution ratios were significantly higher in the 2000s, suggesting the input of combustion and/or anthropogenic PAHs has increased over time in this region.

Foster et al. [91] analyzed the composition and concentrations of hydrocarbons in surface and historical sediments from 11 sites across northern Baffin Bay in eastern Canada prior to oil and gas developments in the area. These provide an assessment of whether recently accumulated sediment (post-1900) differs from older (pre-1900) sediment in this regard. Surficial sediments from northern Baffin Bay that reflected present-day concentrations were found to have hydrocarbon concentrations within a factor of 10 of historic, pre-industrial sediments, indicating that post-industrial anthropogenic sources contribute less than 10% to the natural background levels of PAHs.

Recent (post-2000) studies of temporal trends in PAHs in dated lake sediment cores are limited. Earlier work based on cores collected in the 1990s in Norway and Canada was reviewed by Muir and Rose [95] and is briefly summarized here. Fernández et al. [96,97] measured concentrations and fluxes of 23 PAHs for Lake Arresjøen on northwestern Svalbard (79°40′N) and Øvre Neådalsvatn (62°46′N) in the Caledonian mountain range of central Norway and found that the major PAHs were primarily pyrogenic in origin. Pyrogenic PAH inputs to Arresjøen showed a steady increase from the 1800s with a maximum in top slices [97]. However, Arresjøen had the lowest PAH fluxes of any of the ten European mountain lakes studied. Fernández et al. [96] concluded that the PAH deposition pattern in high altitude mountain lakes, including Arresjøen, paralleled sulfate deposition, pointing to combustion particles as the main input pathway. Rose et al. [19] determined 15 PAHs in surface and pre-historical slices of sediment cores from four lakes on Svalbard. Highest fluxes were found in Lake Tenndammen (360 μg/m^2/y, N.L Rose, University College London, unpubl. data), a lake within 20 km of the coal mining towns of Barentsburg and Longyearbyen. PAH fluxes in the four Svalbard lakes appeared to decline with distance from the Barentsburg/Longyearbyen area.

PAHs (the same 23 PAHs as analyzed by Fernández et al. [96], excluding perylene and retene) were also reported in a series of dated sediment core studies from the Canadian Arctic [98–103]. Total PAH fluxes in southern Yukon lakes ranged from 9.1 μg/m^2/y in remote Kusawa Lake to 174 μg/m^2/y in Little Atlin Lake [103]. Two remote lakes in the Mackenzie River basin, Lac St Therese and Yaya Lake, had much higher PAH fluxes (68 and 140 μg/m^2/y, respectively) [100]. The PAH source for the core from Yaya Lake was thought to be annual inputs from the Mackenzie River during spring floods, with the PAHs themselves probably of natural/petrogenic origin due to crude oil seeps upstream [104]. The historical profiles of 3- and 4-ring pyrogenic PAHs (e.g. fluoranthene, pyrene, retene, BaP) reported for eight Arctic and subarctic lakes increased in concentration from the mid-19th century onward [98–103].

Fluxes of PAHs were determined in dated sediment cores from four lakes in northern Alaska [61] as part of WACAP [58]. The two lakes on the Alaska north slope, Burial Lake and Lake Matcharak, both showed increasing ΣPAH, but different rates of increase (Fig. 8). Lake Matcharak had the highest ΣPAH doubling time (10.5 y) of all sites in the western and northern US lakes studied by Usenko et al. [61]. In contrast, the ΣPAH flux in Wonder Lake in Denali NP, a mountainous region in central Alaska, had a half-life of 15.3 y and a decline was also found in nearby McLeod Lake. As noted previously (Fig. 6), the Alaskan Arctic catchments (Burial Lake and Lake Matcharak) and the Denali catchments (Wonder Lake and McLeod Lake) had significantly different PAH profiles in the sediment suggesting different sources. Using air mass back trajectories, Usenko et al. [61] suggested the PAH sources for Burial Lake and Lake Matcharak could be the Prudhoe Bay oil fields to the northeast. The Prudhoe Bay oil fields are the largest oil fields in North America and began production in 1977. The rise in concentrations in the core from Matcharak appears to fit the increase in crude oil production (Fig. 8).

5. Conclusions

Polycyclic aromatic hydrocarbons (PAHs) may enter the Arctic environment from either natural or anthropogenic sources. Although local pollution inputs from within the Arctic may increase

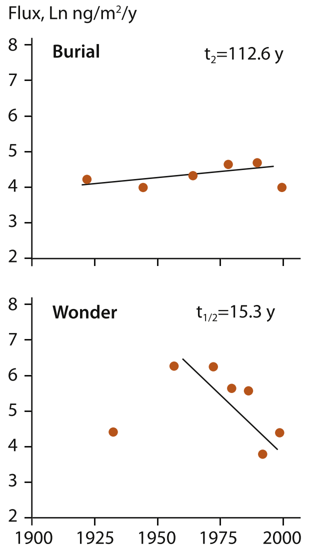

Fig. 8. Focus-corrected flux profiles of PAHs (16 USEPA PAHs and retene) in WACAP sediment cores (natural log (ln) scale). Doubling times (t_2) and half-lives ($t_{1/2}$) are given where least squares regressions were statistically significant ($p < 0.05$) Sources: Usenko et al. [61]; AMAP [14].

in the future as oil exploration and shipping activities increase, they are currently considered to be negligible compared to PAHs atmospherically deposited from the burning of fossil fuels and biomass at lower latitudes.

The numerous potential sources of PAHs make it difficult to attribute trends in their occurrence to specific activities, however, the application of attribution ratios and models have improved understanding of PAH sources and pathways to the Arctic region. Modeling efforts indicate that atmospheric PAHs in the Canadian and Norwegian Arctic are likely to have originated in the circumpolar countries – predominantly Russia, northern Europe, and North America, while mid-latitude East Asia is a minor source, despite contributing more than 50% of global PAH emissions. Models indicate that concentrations and source regions of PAHs in Arctic air can vary seasonally, with the higher concentrations in winter and spring likely to have originated in Western Russia and Europe.

Attribution ratios suggest PAHs found in Arctic marine waters and sediment predominantly originate from natural underwater seeps, while those measured in air, freshwater, and terrestrial environments are likely to have originated from atmospheric and combustion-derived sources. Although data for PAHs in Arctic air are numerous, data for PAHs in terrestrial and freshwater environments remain scarce. Since pyrogenic and biomass combustion sources may be particularly important in terms of PAH concentrations in terrestrial and freshwater environments, determination of a larger suite of PAHs in Arctic air, snow, water, and freshwater sediment samples would provide much needed additional information on sources.

PAHs and their derivatives have been detected in Arctic biota from terrestrial, freshwater and marine environments, indicating their potential to bioaccumulate. Recent modeling studies have suggested that while concentrations of many of the original 12 POPs have declined in Arctic biota over the past 25 years, concentrations of PAHs have risen by as much as 30-fold in fish and mussels, initiating concern over their emerging role in the Arctic ecosystems [13]. However, PAHs may be readily transformed during combustion or from abiotic and biotic processes and thus are not always present in the environment and biota as parent compounds. As PAH derivatives also have the potential to cause adverse effects, monitoring studies of PAHs in Arctic biota should consider including both parent PAHs and their derivatives (e.g., hydroxylated-, nitro- and oxy-PAHs) for more accurate estimates of exposure and toxicity.

Globally, PAH emissions are expected to decline in the future, however models suggest the Arctic may not experience the same magnitude of decline projected for other world regions. Furthermore, future changes in climate may contribute to a re-volatilization of environmental PAHs, providing a source of secondary emissions to the Arctic atmosphere. Thus, future environmental monitoring will be important for understanding the impacts of changing emissions and re-emissions of PAHs in the Arctic.

Conflict of interest statement

The authors certify that they have no affiliations with or involvement in any organization or entity with any financial interest or non-financial interest in the subject matter or materials discussed in this manuscript.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.emcon.2019.04.002.

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