

## Authors' Responses to Reviewers' Comments

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**Manuscript Title:** Polycyclic aromatic hydrocarbons (PAHs) and their nitrated and oxygenated derivatives in the Arctic boundary layer: Seasonal trends and local anthropogenic influence

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We thank the two anonymous referees for their constructive, in-depth comments toward an improvement of the manuscript. Below we provide our point-by-point responses.

### Anonymous Referee #1

#### Reviewer's general comment:

In this study polycyclic aromatic compounds (PACs), including PAHs, as well as nitro- and oxy-PAHs were determined in air in Longyearbyen Svalbard for 8 months. There are many novel aspects to the study. There is a nice demonstration of the effect of polar sunrise in February, where oxy-PAHs begin to be detected. The study also shows that, contrary to reports for remote sites like Zeppelin station at Ny- Ålesund, and Alert in Northern Canada, PAHs are higher in summer due to shipping emissions from high vessel traffic. Local vehicle traffic eg snowmobiles are identified as important sources in the spring (P3) period. Previous studies on PAHs in the Arctic have focused on long range transport and tended to ignore local sources. The interpretation of results also considers back trajectories, wind directions, humidity and the boundary layer height at the sampling site during winter and spring conditions. A large suite of PACs was determined (29 oxy-PAH and 35 nitro-PAHs) making it the most detailed study of these transformation products in the Arctic to date. However, the analysis of unsubstituted and alkylated PAHs was much more limited with the latter group consisting of only 3 methyl-PAHs + retene. Thus source attribution of the PAHs was a bit limited. The use of HPLC-fluorescence, while giving good detection limits, meant that some alkylated PACs would be co-eluting with the target analytes due to more limited peak resolution, resulting in less precise measurements compared with GC-MS (Wise et al. J Chromatog 1993). The authors should acknowledge other possible sources for the oxy-PAHs. Several of the oxy-PAHs are actually industrial chemicals or used in consumer products (eg benzophenone). It is actually quite interesting that benzophenone is detected; possibly it reflects emissions from the sampling site (UNIS) or nearby facilities.

**Author's response:** We would like to thank the Reviewer for his insightful comments toward improving our manuscript. We appreciate all the constructive suggestions and we have carefully addressed it, as explained below.

#### REVIEWER'S SPECIFIC COMMENTS:

Reviewer's comment line 16: Concentration levels can be replaced with "concentrations".

**Author's response:** Corrected accordingly.

line 64: "in summer, the LRAT path to the Arctic shifts to a cleaner one, from the North Atlantic Ocean (Stohl, 2006)".

Reviewer's comment: The discussion here mainly applies to the European arctic. The situation obviously differs across the Arctic eg Canadian arctic archipelago and western Greenland are know to receive LRAT from mid-continental North America as is well documented from air mass trajectories and events such as forest fires (Yu et al ES&T 2019; Keegan et al PNAS 2014).

**Author's response:** Using back trajectories, emission inventories, and atmospheric processing (e.g., photolysis, deposition, etc.), Wang et al. (2010) traced PAH sources to the High Canadian Arctic (Alert). The air pollution at

Alert was dominated by emissions from Russia and Europe in winter and spring (56-85%) as the strong air flow from Eurasia retards the input of pollutants from North America (Wang et al., 2010; Klonecki, 2003). The atmospheric transport of PAHs to the Canadian Arctic was weak in the summer, when the air masses reaching Alert were largely from North Pacific and North Atlantic oceans (Wang et al., 2010; Stohl, 2006). Similar outcome for the Canadian Arctic has earlier been reported by Halsall et al. (1997). Thereby, our description of seasonal pollution delivery to the lower High Arctic troposphere is correct both for the European and the Canadian sectors. However, according to Wang et al. (2010), 90% of total atmospheric concentration levels of PAHs at Alert stems from biomass burning at the Nunavut Territory of Canada and Greenland and the input from islands in northern Canada become more important, which is different from the European Arctic summer sources as fairly indicated by the Reviewer. The text was complimented with an additional information in order to reflect the differences: Line 73: "Thereby, the Arctic front location and permeability, the large scale circulation in the Arctic, available solar radiation, and precipitation regime affect the magnitude of PAC's LRAT to the Arctic lower troposphere, enhancing transport during winter and inhibiting it during summer (Klonecki, 2003; Stohl, 2006; Bozem et al., 2019), when local regional sources become more important and differ across the Arctic (Wang et al., 2010; Stohl, 2006; Keegan et al., 2014; Berthiaume et al., 2020)".

To note, the recent in-depth study by Tevlin et al. (2021), focused on the Canadian ambient air pollution by PACs, reported only episodic contribution of forest fires in Canada, Alaska, and Greenland to the air quality at Alert.

line 115. How is waste handled locally? Landfill only? Could be an important point re sources if incinerated.

**Author's response:** As indicated in text line 115: "There is no local waste incineration". All garbage is collected and shipped to mainland treatment facilities according to the government of Norway regulations since 2001.

line 197. The authors should provide a rationale for their use of HPLC-fluorescence. there are hundreds of PAHs which are difficult to separate even with two-dimensional gas chromatography.

**Author's response:** Quantification of PAHs via HPLC-Fluorescence was performed following the European Committee for Standardization (CEN) standard procedures EN 15549:2008 and TS 16645:2014 (CEN, 2008, 2014) as it stays in the text line 214. Analysis of the NIST SRM1649b provided accurate results ( $\pm 20\%$  except for retene and chrysene) with high precision (RSTD < 10 %) (new Table S8). Ineris has long-term experience analyzing PAHs in ambient air using UHPLC-Fluorescence as it is considered a PAH-sensitive method with very good detection limits and better than those obtained by GC/MS. As in this study we were analysing trace concentration levels it was a major advantage by comparison to GC/MS even if this latter allows the separation and analysis of several useful methyl-PAHs. In addition, (text line 215) Ineris is part of the French national reference laboratory for air quality monitoring (LCSQA) and participates every year/two years (since 2001) to national/European interlaboratory comparison exercises for the analysis of PAHs in ambient air. Results obtained at that period and for the last exercises were in good agreement with reference values and so provide confidence in the method utilization and accuracy.

line 218. The QA data should be provided in the SI eg actual agreement with SRM 1649b. This is particularly important because the data from Zeppelin station were analysed using GC-MS not LC-fluorescence. Would be useful for other researchers if all analytes were reported even those not certified.

**Author's response:** Quantified concentrations of 22 PAHs, 14 nitro-PAHs, and 4 oxy-PAHs in SRM 1649b are now provided in the SI as Table S8. They are compared to certified, reference or indicative concentration values available in the NIST certificate of analysis. Concentration values for other PAH derivatives have been reported previously in Albinet et al. (2006) and Albinet et al. (2014).

line 258: "The measured PAH concentrations in Longyearbyen were similar (about 19% higher) to those detected at the Zeppelin station (Table S7), confirming LRAT of anthropogenic pollutants as a dominant source of PAHs in the Arctic air during winter".

**Reviewer's comment:** Are results comparable given the different methods of analysis. Is this for the sum of the same analytes. Similarly the comparison with Pallas depends on same analytes and method performance although the samples from Pallas were analysed by LC-fluorescence

**Author's response:** The sums of the same PAHs (different suits for Pallas and Zeppelin) were compared. This information is outlined as footnotes to the Table S7. The methods appeared to differ for UNIS (UHPLC-Fluorescence) vs Zeppelin (GC-MS) and were similar for the UNIS vs Pallas (LC-Fluorescence) data.

line 356: "The summer PAH levels are usually 1-2 orders of magnitude lower than in winter and often close to detection limits (Table S7 and Yu et al., 2019; Prevedouros et al., 2004; Singh et al., 2017). Nonetheless, summer  $\Sigma 22$  PAHs concentrations ( $1779 \pm 1210$  pg m<sup>-3</sup>) measured in Longyearbyen air were about 67% higher than the winter P1 levels (Table S1)".

**Reviewer's comment:** Yu et al (2019) summarized the cold and warm season concentrations and differences vary greatly among individual PAHs. In their summary, fluorene had the greatest difference (8-fold higher in cold season). The median difference for average concentrations of 19 PAHs was 3.4. Thus the statements here re order of magnitude need more precision.

**Author's response:** Additional information have been provided in the text (red colored):

"summer  $\Sigma 22$  PAHs concentrations ( $1779 \pm 1210$  pg m<sup>-3</sup>) measured in Longyearbyen air were about 67% higher than the winter P1 levels (Table S1). For individual PAHs, largest differences ( $\geq 2$  times higher concentrations in summer) were observed for phenanthrene, 2-methylnaphthalene, acenaphthene, pyrene and chrysene while other compounds showed comparable concentrations in both seasons".

line 417: Spelling methylnaphthalene

**Author's response:** Corrected accordingly.

line 438: "Comparing the PAHs spring weekend profiles (n = 7) with the rest of the cold period day profiles (P1-P3; n = 12) confirmed a significant increase of benzo[a]anthracene proportion caused by snowmobiling, as well as the higher contribution of 1-methylnaphthalene, fluoranthene, acenaphthene, and coronene to the total PAHs profile (Fig. S7). All over, benzo[a]anthracene, phenanthrene, and 1-methylnaphthalene accounted for 68 % of the average PAHs profile of these days with known high snowmobile traffic. The predominance of phenanthrene and methylated PAHs in snow samples contaminated by a snowmobile emission has being reported earlier (McDaniel and Zielinska, 2014), although the contribution of 2-methylnaphthalene and 2-methylfluoranthene in the present study was found negligible. Different engine technologies and fuel quality may have been responsible for it".

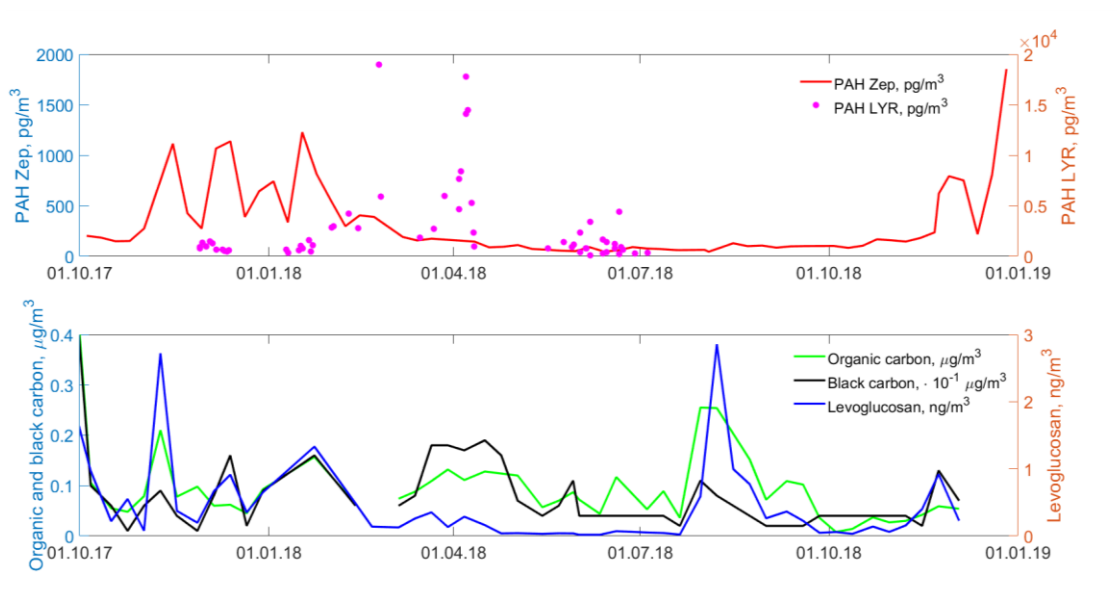
**Reviewer's comment:** Unfortunately, this study did not look at alkylated PAHs in depth and thus conclusions about fuel and engine emission sources are very uncertain.

**Author's response:** Yes, we did not measure key methyl- or alkylated PAHs that would have been useful to make such discussions. However, the gasoline engine emission influence was indicated by the 3 times higher values of (BghiP+Cor)/ $\Sigma$ PAH<sub>p</sub> diagnostic ratio values in spring (Fig. 2). The detailed analysis of the PAH profiles of the days with known high snowmobile traffic revealed increased profile proportion of 1-methylnaphthalene as indicated in the main text line 434. Although the 2-methylnaphthalene and 2-methylfluoranthene concentrations increased by factors of 1.7 and 2.7 on these days, respectively, their proportion in the total 22 PAH profile did not rise, which means that the rate emissions of other PACs listed in the text were higher (Fig. S7). Consequently, 2-methylnaphthalene and 2-methylfluoranthene were not indicated as specific to the snowmobile emissions PAHs. However, higher concentrations of 2-methylnaphthalene and 2-methylfluoranthene in the twilight and spring periods compared to the winter period (Table S1).

Table S1. What about retene? It is not discussed in the main text but was determined in all samples. As a component of coal dust and biomass burning, but not from combustion of fossil fuels – it could be helpful to identify sources

**Author's response:** The retene ambient concentrations were quite stable being only 20% higher in spring (the seasonal concentrations plot is provided below and Table S1), when the local coal-burning power plant emissions were downwind the UNIS sampling site all the time. The concentrations of all the individual PAHs increased by a factor of 4 as average in spring (Table S1). Our study shows that this was mainly caused by accumulation of near-ground emissions in the local boundary layer (lowest 200 m) under stably stratified, sustained cold conditions with deep thermal inversion in spring as illustrated on Fig.2 and discussed in the text line 299. In these conditions the LRAT is minimal.

We are aware that the PAH levels in Svalbard air can be significantly affected by forest fires in Russia (Luo et al., 2020), though we believe that these emission plumes were not caught during the air sampling in Longyearbyen as retene concentrations did not peak significantly and both the levels of PAHs (including retene) and levoglucosan, the key molecular marker of biomass burning (Simoneit et al., 1999; Bhattarai et al., 2019), were low in spring in Ny-Ålesund (the plot is provided below). In any case, only the use of levoglucosan would have permitted to obtain a robust conclusion on the impact of biomass burning as retene is not highly specific of this combustion source.



line 454: “Benzophenone, phthalic anhydride, 9-fluorenone, 1-naphthaldehyde, and 1,2-naphthalic anhydride were the predominant substances and accounted for  $89 \pm 4$  % of the total oxy-PAHs in all periods (Fig. 5b). Main changes in the profiles were caused by a 16 % increased contribution of benzophenone in spring and a 17 % higher proportion of phthalic anhydride in summer. Among the minor compounds (ranging from 25 to 400 pg m<sup>-3</sup>) accounted for about 10 % of the total oxy-PAHs profile, higher proportions of 2,3-naphthalenedicarboxylic anhydride and anthrone were found in summer. Detailed analysis of the average summer profiles based on different prevailing wind directions (Fig. S9) did not reveal any primary local source influence. This may indicate the secondary formation of phthalic anhydride, 2,3-naphthalenedicarboxylic anhydride, and anthrone from PAH photochemical reactions (Keyte et al., 2013; Chan et al., 2009; Perraudin et al., 2007; Bunce et al., 1997; Lee and Lane, 2009)”.

**Reviewer's comment:** Benzophenone is an industrial compound eg used as a UV blocker in plastics. It is not really a PAH oxidation product unless its precursor diphenylmethane is included. Looks like the sampling is picking up a lot of emissions from products rather than from combustion. Similarly phthalic anhydride is probably from plastics rather than atmospheric oxidation of benzofuran.

**Author's response:** We did not state the secondary origin of benzophenone. The ambient concentrations of benzophenone increased a factor of 3 in spring compared to winter (Table S2) and the oxy-PAHs chemical profile was significantly shifted by this compound (Fig. S10). Thereby, benzophenone was likely emitted from

snowmobiles (text line 467). No plastic burning, or emission from plastic industries, are expected in Longyearbyen, though snowmobile drive belts might burn when high speed accelerated and at the cold start. The phthalic anhydride is known product of atmospheric oxidation of naphthalene, 2-methylnaphthalene, and phthalaldehyde (Chan et al., 2009; Keyte et al., 2013). The subject of the PAH chemical processes and secondary formation of oxy- and nitro-PAHs in the polar troposphere will be comprehensively addressed in the forthcoming paper (in preparation) based on the suit of results presented in the current work together with results obtained at other locations in Svalbard.

## Anonymous Referee #2

### Reviewer's general comment:

This manuscript investigated the seasonal variations and long-range transportation of polycyclic aromatic hydrocarbons (PAHs) and their nitrated and oxygenated derivatives on a remote island in the European Arctic. Considering the importance of the topic, it is a significant contribution to fully understand the current contamination status with both local emissions and those from midlatitudes via different transport processes. The article is generally well-written and derives reasonable conclusions. Hence, I recommend it for publication in the journal after some revision.

**Author's response:** We appreciate the Reviewer's careful consideration of our manuscript and constructive comments. We have addressed the reviewer's specific comments below.

### REVIEWER'S SPECIFIC COMMENTS:

L105-140: a high-volume sampler was deployed on the roof of UNIS for air sampling. In such case, the local emissions might be highly controlled by the cars or snowmobiles parked in front of UNIS. The contributions from the plumes of the harbor and the power station of Longyearbyen might be only remarkable in a few samples. Apart from winter, it can be very hard to identify the proportion of PAHs originated from midlatitudes owing to many removal processes on the way to Longyearbyen.

**Author's response:** The reviewer assumes correctly. The LRAT prevailed during winter, while mainly local anthropogenic emissions contributed to the spring and summer ambient concentrations of PACs.

The sampling site description is outlined (line 122) as "Considering south-easterly prevailing wind direction and katabatic winds from Longyearbreen and Larsbreen glaciers, nearly all year-round UNIS is located downwind from the town (600 m distance), and the two main gateways (Adventdalen and Longyeardalen valleys) largely used for snowmobile driving during winter and spring. In summer, the wind direction can temporarily change to northwesterly (Dekhtyareva et al., 2016; Niedźwiedz, 2013), making UNIS position downwind the coal-burning power plant (1 km distance to UNIS) and the harbour (5 km distance to UNIS). UNIS is also surrounded by paved roads that are used by cars all year round (Fig. S1c)".

We have considered the contribution of the harbor and the power station emissions only during summer, on the days with NW wind (line 366 and 392; n=7).

L197-200: For instrumental analysis, GC-NICI-MS was used to quantify 35 nitro-PAHs and 29 oxy-200 PAHs. If a GC-MS is available, what is the advantage to analyze PAHs by UHPLC-Fluorescence. It can add additional uncertainty for comparison with PAH data from Zeppelin station.

**Author's response:** Quantification of PAHs via UHPLC-Fluorescence was performed following the European Committee for Standardization (CEN) standard procedures EN 15549:2008 and TS 16645:2014 (CEN, 2008, 2014) as it stays in the text line 214. Analysis of the NIST SRM1649b provided accurate results ( $\pm 20\%$  except for retene and chrysene) with high precision (RSTD < 10%) (new Table S8). Ineris has long-term experience analyzing PAHs in ambient air using UHPLC-Fluorescence as it is considered a PAH-sensitive method with very good detection limits and better than those obtained by GC/MS. As in this study we were analysing trace concentration levels it was a major advantage by comparison to GC/MS even if this latter allows the separation and analysis of several useful methyl-PAHs. In addition, (text line 215) Ineris is part of the French national reference laboratory for air

quality monitoring (LCSQA) and participates every year/two years (since 2001) to national/European interlaboratory comparison exercises for the analysis of PAHs in ambient air. Results obtained at that period and for the last exercises were in good agreement with reference values and so provide confidence in the method utilization and accuracy.

L215-208: The high field blank contamination of biphenyl-2,2'-dicarboxaldehyde, 2,3-naphthalenedicarboxylic anhydride, and 1,8-naphthalic anhydride present in PUF samples. Actually, these chemicals are not only occurring in nature, but also synthesized for industrial application. Thus, the blanks might be from the PUF matrices.

**Author's response:** These compounds were not detected in laboratory blanks. Thus, we concluded that the samples were contaminated during sample collection and handling.

L210-212: the LOQ is useful to control the data quality for the compounds which are not detectable in field blanks. While for the compounds present in field blanks, it might be good to calculate the method detection limit with field blanks.

**Author's response:** The MDL based on field blanks' contamination was applied to the samples with a critical value of 30% of the seasonal average PAC's concentration (line 206). In other words,  $MDL = FB + 0.3 * X$ , where FB is an average field blank value (QFF or PUF; n=8) and X is the compound seasonal mean concentration value.

L235: Fig.1 shows the concentrations of Oxy-PAHs are 2-10 times higher than their parent PAHs. Are the Oxy-PAHs determined in this work mainly from LRAT or resulted from degradation of local PAHs?

**Author's response:** Based on the intercontinental transport regimes' and local meteorology conditions' investigations, it was concluded that the polluted air masses were effectively transported to Svalbard during winter, while local emissions were the primary air pollution sources in spring and summer. Oxy-PAHs are emitted from the same primary sources as PAHs and can be complimentary produced in the atmosphere during northward LRAT or within the Arctic region. It is not easy to distinguish the predominant sources (primary vs secondary) and the emission and production rates vary between the compounds. The subject of the PAH chemical processes and secondary formation of oxy- and nitro-PAHs in the polar troposphere will be comprehensively addressed in the forthcoming paper (in preparation) based on the suit of results presented in the current work together with results obtained at other locations in Svalbard.

L425: Fig.5b shows five oxy-PAHs accounted ca. 90% of the total oxy-PAHs in all seasons, how is about their parent PAHs?

**Author's response:** Benzophenone (BZPh), phthalic anhydride (PhtAn), 9-fluorenone (9-Flu), 1-naphthaldehyde (1-NapA), 1,2-naphthalic anhydride (1,2-NapAn) accounted for almost 90% of the total oxy-PAHs as correctly noted by the Reviewer. These compounds can be primary emitted and secondary formed in the atmosphere. In the forthcoming paper, we will evaluate the secondary origin of these oxy-PAHs. The following generation pathways of the listed oxy-PAHs were considered: BZPh/Ant, PhtAn/Nap, PhtAn/Ant, PhtAn/PhtA, PhtAn/2-MeNap, 9-Fluo/Flu, 9-Fluo/Phe, 9-Fluo/Ant, 1-NapA/Nap, 1-NapA/1-MeNap, 1,2-NapAn/Phe, 1,2-NapAn/Ant. Among the potential parent PACs, Nap, Phe, Flu, and 1-MeNap were predominant PAHs detected in the current study (Table S1 and Fig. 5a), while concentrations of Ant, PhtA, and 2-MeNap were notably lower, which might potentially indicate their lower primary emissions or/and significant photochemical degradation and transformation into oxy-PAHs. However, this topic is outside of this paper's scope and full details will be provided in the follow up paper (in preparation).

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