Response to anonymous reviewer #1

We thank the reviewer for their careful consideration of our manuscript and their helpful comments.

We address the review comments (blue text) point-by-point as follows:

Overview of the obtained results (section 5) has several scientific problems, as outlined below in more detail, and these problems need to be fixed before the paper can be accepted for publication. Concerning section 6, I am not fully in favor of putting strategic/political (section 6.1) and scientific (sections 6.2 and 6.3) aims side by side, but it is up to the editor to decide whether this requires some restructuring of the text.

We address the points raised for Section 5 in the remaining comments below. The discussion on strategic goals has been moved to a separate section 7 to distinguish between political and scientific issues.

Section 5.2.

I do not understand the first statement of section 5.1 (lines 652-653). Based on figure 6, it is impossible to see whether OC and EC resemble each other at any time (their concentrations levels certainly do not, and resemblance of concentrations ratios is also questionable). Overall, I do not see how Figure 6 could tell anything about the similarities in OC and EC source regions.

We have deleted the first statement of section 5.1 as this requires a more thorough discussion of a specific issue which is then beyond the broad scope of our article. To show that the time series of OC and EC resemble each other, we provide a scatter plot in a second panel in Figure 6, showing a Pearson's R^2 correlation of 0.96 in October to May 2019, and 0.77 for June to September 2019.

The statement on lines 575-677 is unclear. I suppose that the authors mean that the CF conversion factor typical for aged aerosols should be applicable for Zeppelin because of its remote location from main sources. The wording (complies well with) gives an impression that this thing has somehow confirmed for Zeppelin.

The conversion factor we use is that suggested for non-urban sites by Turpin and Lim, 2001. We have changed the text to "A conversion factor (CF) of 1.9 to 2.2 is suggested for conversion of OC to organic matter (OM) for non-urban aerosol (Turpin and Lim, 2001), such as at the Zeppelin Observatory, where most aerosol particles are long-range transported."

Russian is a very large territory. Do the authors have more detailed information on the main source areas for high sulfur episodes, e.g. the Kola Peninsula area discussed a lot in previous literature?

Indeed, the Kola Peninsula (particularly smelters in the area) is a known major source, also discussed in the original reference by Aas et al. This is included in the revised manuscript: "These episodes of sulfur pollution occur due to the arrival of air masses from Russia, for example the Kola Peninsula, due to the presence of heavy industry including non-ferrous metal smelters (Aas et al., 2020)."

When discussing about past trends of inorganic ion concentrations in atmospheric aerosols (lines 686-696), I wonder why the authors refer to targeted emission reductions during 1990-2010, not the real emission reduction that took place. Data on actual emission reductions during that period is certainly available.

The next statement in the manuscript is in fact a comparison to the real-world emissions. For clarity, we specify this in the revised manuscript as follows "This is significantly lower than the real-world

reductions seen on the Norwegian mainland, e.g. SO_2: 95%, sea salt corrected SO_4^{2-}: 74% at Birkenes, South Norway (Aas et al., 2020),"
It is important to note that these inorganic ion measurements are connected to the need to monitor these species in the GP, and so the progress in real-world compared to the targeted emissions is highly relevant.

Section 5.2.

This section is about aerosol physical and optical properties (the word optical could be included into the title), so why do the author start the section by mentioning CCN and cloud properties which are not discussed at all in this section?

We note that several of the papers covering aerosol physical properties in the Arctic are linked to the influence of such properties on CCN, but that this does not need to be explicitly stated here. For clarity we have changed the opening sentence to "There are large number of atmospheric aerosol and cloud studies in polar regions based on short term campaigns, carried out predominantly in the polar day season."

lines 702-704: The authors assume implicitly here that the particle size is some sort of proxy for its ageing. This is probably true but should be explained for readers not familiar with combined effects of aerosol sources and aerosol dynamics taking place during atmospheric transportation.

A clarification of what is observed is added, as well as an explanation of the processes driving growth in these particles, which is commonly called ageing. We state this explicitly in the revised manuscript as follows: "These particles, when observed in remote areas, are usually formed through atmospheric processes such as condensation, coagulation and cloud processing, and are commonly referred to as 'aged' particles (See Fig. 7). "

Please explain in more detail what is meant by "light period" and "summer" (lines 710 and 711), and whether "sunlit period" (line 724) means something else.

The text has been revised to clarify that nucleation is highest during the lightest periods of the summer, specified as follows: "Particle nucleation events followed by particle growth are observed frequently during the lightest part of the year, i.e. April to September, and dominate the particle concentration during the summer, i.e. June to August (see Fig. 8 and Tunved et al., 2013)"

The discussion about aerosol optical properties is vague (lines 732-745). The authors try to relate changes in optical properties to those in particle concentrations (number or mass, not explained?), but the relevance or purpose of this exercise has not been explained. I do not understand what the authors mean be stating the particle concentrations increase through the year (line 740). This whole paragraph needs to be rewritten.

It is a misunderstanding that we try to relate the optical properties to the size distributions given in figure 7 to 9. It is not written anywhere. In this paper we only give examples on how different types of observations have given a better understanding of the Arctic atmosphere and its composition. The optical observations are made over 18 years to compare with 10 years for the particle size measurements supporting a more rigid trend analysis. There is no such trend analysis presented for the particle size measurements yet. Thus, no comparison is made. However, we have rewritten the section on the optical properties to make it as clear as possible. A more detailed explanation on what influences the optical properties has been added: "Recently, 18 years of nephelometer measurements at the Zeppelin Observatory were used to evaluate the trends of particle light scattering properties

(see Fig. 10 and Heslin-Rees et al. 2020). An increase in particle light scattering indicates either an increase of particle concentrations or an increase in particle size; the latter is supported by a decreasing scattering Ångström exponent, showing a shift to larger particles in the particle size distribution. The increase in particle size and particle light scattering coefficient seen throughout the 18 years most likely corresponds to an increased contribution from larger particles such as sea spray. Heslin-Rees et al. (2020) argue the observed long-term changes are due to changes in atmospheric circulation, i.e., an increased frequency of long-range transport from the open northern Atlantic. However, new particle formation (NPF) events at the Zeppelin Observatory have been shown to be anti-correlated with sea ice extent indicating a dependence on more open sea Dall'Osto et al. (2017). This is also supported by number of recent studies linking ocean biological activity with biogenic sulfur variability and abundance in the Arctic atmosphere (Jang et al, 2021) and related aerosol properties and cloud condensational nuclei variability (Choi et al, 2019, Park et al, 2021). Naturally driven NPF dominates the summertime Arctic atmospheric aerosol, even though the detailed physiochemical process pathway is not known and is a subject of ongoing research."

The discussion on lines 755-764 is rather general and appears to be loosely connected with other contents of section 5.2.

This section belongs in the aerosol measurement methodology in 4.2 and has been moved accordingly

Section 5.3

What is the point of bringing up CO2 concentration in 2019 and its increase from the previous year? The CO2 increase is a well-known fact, while its annual increase rate varies from year to year. Data from one single year provide little insight on this matter (lines 766-768).

The comparison to the previous year has been removed and the sentence is now "CO_2 is increasing with a long-term trend of 2.5 ppm per year (Table 5) and has increased by ≈15% since 1989 levels (357 ppm)."

What is the basis for stating that the CO2 concentration increase rata is exponential? (line769)

This statement has been rephrased: "It should be noted that the growth is positive in all years, highlighting the challenge in meeting emissions reductions needed to meet the Paris Agreement goal of keeping the global annual average temperature increases below 2℃."

line 793: any explanation for the stated pause of CH4 mixing ratio?

The reason for this pause is discussed in detail in the section that immediately follows and we do not feel that any further detail is needed.

There is repetition of text between the lines 838-844 and lines 858-869. Also figure 12 appears twice in the paper.

The repetitions in text and the extra figure have been deleted in the revised manuscript at the second occurrence.

Section 5.6:

Based on measurements of just one site in Arctic and one site outside Arctic,it is impossible to make any general statement about differences between Article areas

and those outside Arctic (lines 992-994).

This general statement has been removed from the main text.

Section 6.2:

There is much new scientific work and findings on arctic amplification and related issues that seem to be missing in the introductory part of this section (lines

A thorough review of Arctic amplification is beyond the scope of this article. Meanwhile, the observation of rapid Arctic warming alone, rather than a discussion of its causes, is more relevant to the section. Hence, we have removed the discussion of Arctic amplification from the revised manuscript.

Technical and minor scientific issues

line 227: something is missing from here (e.g. … during 1971 to 1980)

'From' has been added. "This conclusion was based on extensive climatological tabulations of the meteorological observations in Ny-Ålesund from 1971 to 1980."

line 435: INP should be in parenthesis

This has been corrected in the revised manuscript.

line 440 (and later line 765): The term "climate gases" is not commonly in use. Please consider modifying the titles.

We have modified this to the more explicit "Atmospheric trace gases of high relevance to global climate change"

line 711: Figure 8 does not tell anything about nucleation and particle growth, so it should not be referred to here but later in the text.

Particle growth and nucleation directly impact particle mass, and these are the main causes of seasonal variability in Fig. 8. We do not feel a change is needed here.

line 795: is it possible to measure the CH4 concentration with a 5-digit accuracy?

It is indeed possible to measure with such precision: the G2401 manual states that the precision is at least +/- 0.3 ppb for 1 hour averaging (we actually average over longer for the daily means). See https://www.picarro.com/support/library/documents/g2401_analyzer_datasheet

line 801: an increase from 28 to 32 is not consistent with 25% increase.

This has been deleted in the revised manuscript.

lines 811 and 815: suggest à suggested

Since the change is recent and ongoing (at least until 2020) these statements seem grammatically correct with respect to the use of present and present continuous tenses.

line 1008: MC or MCM?

This has been changed to MCM at the first instance

lines 1013-1014: it is enough to explain GEM one time.

This has been corrected in the revised manuscript.

line 1037: … Asia, including China, contribute…

The comma has been added to the revised manuscript

line 1092 vs. line 1143: please use only a single term for LOE (episodes of very low ozone vs. low ozone episodes).

We have opted to use 'low ozone episodes' for consistency

lines 1119-1121: Unclear sentence, please modify.

We have split this sentence in the revised manuscript: "Furthermore, links between climate change in other regions and the frequency of Arctic LOEs have been proposed. For example. (Koo et al., 2014) found correlations between so-called 'teleconnection patterns', i.e. weather patterns in other regions, such as the Western Pacific, and the frequency of LOEs in the Arctic."

lines 1166-1168: the first sentence of section 6 is unclear. Please re-write.

We have changed this to "The main focus of atmospheric research has shifted over the decades."

line 1184: please correct the grammar (that need study)

This has been changed to "requiring study".

line 1125: a paper in preparation is not a proper reference.

This reference and accompanying statement have been removed for the revised manuscript. Instead, we provide a broader statement with a finished paper reference: "A shift in the natural aerosol baseline within the Arctic is evident and an improved knowledge of the individual processes is needed to better constrain the future development of the Arctic climate (Schmale et al. 2021)."

line 1316: please correct the grammar (will need study)

This has been changed to "require study".

lines 1332 and 1334: CEAC or CEC?

CEAC has been changed to "CECs in the Arctic" for the revised manuscript.

lines 1363-1364: unclear sentence, please modify

This has been corrected to "While the observatory at its inception was primarily focused on national monitoring, the Zeppelin Observatory now hosts measurements from 17 institutions in 13 countries.

Although these are mostly long-term measurements, the Zeppelin Observatory regularly hosts instruments for short-term (1 to 3 years) campaigns. Measurement capabilities have been continuously improved to include state-of-the-art instrumentation."

Some of the figures (figs. 13, 14 and 20) are of poor technical quality.

We have improved the figure quality for the revised manuscript.

Figures 12 and 18 appear twice in the text.

Duplicate figures have been removed from the revised manuscript.