

We would like to thank the Reviewer 2 for useful comments that allowed to improve the manuscript's quality.

In addition to corrections performed according to comments from the Reviewer 1 and 2, the following major changes in the manuscript have been introduced:

- 1) Following suggestion of the Reviewer 2, the description of O<sub>3</sub> profiles has been moved to the beginning of the Results section and Figure 10 became Figure 4 in the new manuscript, and numbering of the other figures has been modified accordingly.
- 2) In the Discussion (section 4), the results from 2017 campaign are contrasted to modelled NO<sub>2</sub>/NO<sub>x</sub> ratio and NO<sub>2</sub> produced through PAN decomposition, and long-term observations from Ny-Ålesund, weather regime and trajectory data are utilized to confirm the large-scale circulation and air pollution links.
- 3) Four new plots have been added in the Appendix and discussed in the Discussion section.
- 4) The Conclusion section is revised to reflect the changes introduced in the manuscript.

### **Reply to the major comments**

1) We agree that the lack of NO<sub>x</sub>/O<sub>3</sub> instrument co-location in Adventdalen and Ny-Ålesund during the 2017 campaign is a major drawback of this study. The NO<sub>x</sub> monitoring in Ny-Ålesund is a long-term ongoing air quality project, and relocation of the instrument from the village to the Zeppelin station would introduce bias in the long-term observations in the settlement. The study in Adventdalen was the first combined air pollution and meteorological field work in Longyearbyen. The measurements there were done by the main author, and only NO<sub>x</sub> monitor was installed there due to the limited grant funding.

We would like to specify that the meteorological data from Ny-Ålesund were not missing. The data from the meteorological station operated by the Norwegian meteorological institute located 100 m away from the monitor have been used in combination with NO<sub>x</sub> data from the village (p. 5 line 120 in the manuscript). The O<sub>3</sub> and CO data from the Zeppelin station have been combined with the meteorological data from the Zeppelin station.

Regarding NO<sub>x</sub> and O<sub>3</sub> data from Ny-Ålesund, the authors have been in contact with atmospheric scientists from Ny-Ålesund research community, and, to our knowledge, the only collocated NO<sub>x</sub> and O<sub>3</sub> measurements from this settlement were performed at the Zeppelin station from February to May 1994. The results were published in Beine et al., 1996. In that study, the combination of NO<sub>x</sub> data and concentration of particles with diameter below 10 nm, atmospheric stability and wind direction was used to identify possible local pollution events. In spring 1994, the local pollution was detected at the Zeppelin station during 6.4 % of the measurement time, and the number of these events was increasing with increased insolation in May. Following this method of event detection, the concentration of particles with diameter of 10 nm routinely measured by DMPS at the Zeppelin station and threshold of > 95 percentile have been used to identify peaks in concentration of newly formed particles. Similarly to the results of Beine et al., 1996, the peak events were detected at the Zeppelin station only in the second part of the 2017 measurement campaign (from 24<sup>th</sup> of April to 13<sup>th</sup> of May). The northerly wind direction was present only during 12 out of 45 hours with peak particle concentration, however, none of these cases was characterized by increased CO concentration at the Zeppelin station. Thus, these peaks in concentration of small particles might have been related to natural rather than anthropogenic emission sources. Indeed, Heintzenberg et al., 2017 described the offset in new particle formation towards late spring and summer when biological emissions become important sources for this process. Therefore, both statistical comparison of the O<sub>3</sub> concentrations in clean and potentially polluted air masses mentioned in the manuscript and absence of coinciding peaks in particle

concentration and CO concentration indicate that the O<sub>3</sub> observations at the Zeppelin station were not significantly affected by local NO<sub>x</sub> pollution during 2017 campaign. Therefore, the lines 226-244 in the section 2.5 have been rewritten and the results about influence of local NO<sub>x</sub> pollution on the O<sub>3</sub> concentrations at the Zeppelin station have been modified accordingly (lines 288-305 in the new version of the manuscript).

2) We agree with the Reviewer about this point, and as it is stated above, the lines 226-244 in the section 2.5 have been rewritten and the results about influence of local NO<sub>x</sub> pollution on the O<sub>3</sub> concentrations at the Zeppelin station have been modified accordingly (lines 288-305 in the new version of the manuscript).

3) The sentences have been modified as following (lines 271-276 in the new manuscript):  
Low correlation between the NO<sub>x</sub> values at the three stations indicates the importance of local emission sources and micrometeorology (wind channelling and spatial variation in atmospheric stability) rather than synoptic meteorological conditions. The background NO<sub>x</sub> concentrations observed in Svalbard in previous studies (Beine et al., 1997) using different measurement techniques are below 0.4 ppb, and thus, the natural variability in NO<sub>x</sub> values due to long-range transport to Svalbard would be undetected in the NO<sub>x</sub> datasets presented in the current study.

4) a) the word “strong” has been changed to “moderate”; b) as mentioned above, the lines 226-244, 271-276 and 288-305 in the new version of the manuscript have been rewritten.

5) The standard error of the mean has been included in the Figure 5 (former Figure 4) for each hour. To improve readability of the figure, separate subplots a) and b) have been made for NO<sub>x</sub> and O<sub>3</sub> data, and supporting text has been modified accordingly. As noted by the Reviewer 2, the Zeppelin station is located at the altitude of 474 m a.s.l. and mostly samples air from the free troposphere with higher O<sub>3</sub> concentration, and thus the data from this station does not exhibit similar diurnal variation as the Barentsburg station. However, the magnitude of these effects is small, and according to the t-test and the WRS-test, there is no statistically significant difference between the nighttime and daytime O<sub>3</sub> concentrations measured at the stations (Table 1).

6) Following recommendations of the Reviewer 1, we included investigation of possible downward transport of O<sub>3</sub> enriched air masses in the trajectory analysis and added the percentage of trajectories descending from higher altitudes (>2000 m) (Figure 9 in the new manuscript). As in previous studies of Hirdman et al. 2009, the downward transport of O<sub>3</sub>-enriched air masses from higher altitudes played significant role during the 2017 campaign. The percentage of trajectory points reaching elevations above 2000 m was highest for the sub-periods III, VI and IX (27%, 33% and 24% of the total number trajectory points for each sub-period respectively). In contrast, during the sub-period VIII with the lowest O<sub>3</sub> concentration at both stations, the percentage of elevated trajectory points was minimal, only 4%. One can also see that the percentage of elevated trajectories varies for the same type of weather regime and determines importance of the downward air mass transport for the measured surface O<sub>3</sub> concentrations in different sub-periods (e.g. ScTr regime in Figure 8c) and e) and Table2). (lines 452-455).

The discussion about NO<sub>x</sub> transport and PAN decomposition modelling has been included in the Discussion part of the paper (lines 501-522).

7) Indeed, there is a difference between the O<sub>3</sub> levels at the Zeppelin station and in Barentsburg as revealed in the radisonde data, but moderate correlation is still valid as both sites are influenced by the same long-range transport events. The analysis of long-term data from the Zeppelin station included in

the Discussion part of the new version of the manuscript (lines 566-610), allows to investigate influence of different weather regimes and obtain more robust results.

As suggested by the Reviewer 2, the Figure 10 (Figure 4 in the new manuscript) and discussion about the vertical differences is moved in the beginning of the Results part, before discussion about diurnal variation and Figure 5.

8) To investigate the influence of local NO<sub>x</sub> emissions on the O<sub>3</sub> concentration in Ny-Ålesund, the following part has been included in the Discussion part of the paper (lines 529-543).

To investigate the influence of local NO<sub>x</sub> emissions on the O<sub>3</sub> concentration in Ny-Ålesund, as it is required in the third hypothesis stated in the introduction of the current paper, we may use historical observations. The data from only six O<sub>3</sub> sonde launches were available for the 2017 campaign (Figure 4). However, the long-term data below 100 m from the O<sub>3</sub> sonde profiles may be used to study influence of the local NO<sub>x</sub> pollution in Ny-Ålesund on the O<sub>3</sub> concentration. These observations are suitable for this purpose because the O<sub>3</sub> sonde launching facility is located just 200 m to the south-south-west and 500 m to the south from the NO<sub>x</sub> monitor and diesel power plant, respectively. Thus, when the monitor detected NO<sub>x</sub> concentration above long-term springtime average in the launch hour, the influence of locally polluted air masses might have been observed in the lowest O<sub>3</sub> sonde data. There were in total 59 O<sub>3</sub> sonde launches, for which NO<sub>x</sub> monitor data was available in spring 2009, 2010, 2015, 2016, 2017 and 2018. The O<sub>3</sub> profile data in the lowest 100 m have been extracted for all 59 launches and grouped according to the NO<sub>x</sub> concentration detected by the monitor and wind direction in the O<sub>3</sub> sonde profiles: 1) above mean NO<sub>x</sub> concentration and northerly wind direction; 2) below or equal to mean NO<sub>x</sub> concentration and northerly wind direction. The median and mean O<sub>3</sub> values below 100 m in the group where the NO<sub>x</sub> values were above NO<sub>x</sub> mean were 11 % and 15% lower, respectively, than for the second group with northerly winds, but without elevated NO<sub>x</sub> concentration. Thus, the O<sub>3</sub> concentration in lowest 100 m downwind from the power plant in the settlement may be reduced significantly due to local NO<sub>x</sub> emissions, but the frequency of such events is unknown in absence of continuous O<sub>3</sub> measurements in the village.

9) The sentence has been modified as following (lines 636-367):

In contrast to NO<sub>x</sub>, the concentrations of O<sub>3</sub> in Barentsburg and at the Zeppelin observatory are moderately correlated and depend on synoptic conditions that promote transport of air masses enriched or depleted in O<sub>3</sub>. In other words, both these stations are regionally representative for the O<sub>3</sub> concentrations.

We would like to thank Reviewer 2 for the suggestion of the alternative NO<sub>x</sub> measurement techniques. We agree that instruments used in this study are more suitable for urban air pollution studies, however, we decided to install the chemiluminescence analyser for the 2017 campaign in Adventdalen to make measurements comparable with data of the same type from Ny-Ålesund and Barentsburg and to take advantage on the available long-term NO<sub>x</sub> data from Ny-Ålesund that is included in the new version of the manuscript.

We will definitely consider suggested measurement techniques for background NO<sub>x</sub> observations during planning of new field campaigns in the Arctic.

### **Reply to the minor comment**

The sentence has been modified as following (lines 450-451 in the new manuscript):

In the sub-period II, 67% of the data from the Zeppelin station were missing (Figure 2). The O<sub>3</sub> concentration in Barentsburg was above median for this sub-period, and the trajectory data show the air masses arriving from the south-east (Figure 9b).

## References

- Beine, H. J., M. Engardt, D. A. Jaffe, Ø. Hov, K. Holmén, and F. Stordal. 1996. "Measurements of NO<sub>x</sub> and Aerosol Particles at the Ny-Ålesund Zeppelin Mountain Station on Svalbard: Influence of Regional and Local Pollution Sources." *Atmospheric Environment* 30(7):1067–79.
- Beine, H. J., D. A. Jaffe, J. A. Herring, J. A. Kelley, T. Krognes, and F. Stordal. 1997. "High-Latitude Springtime Photochemistry . Part I : NO<sub>x</sub> , PAN and Ozone Relationships." *Journal of Atmospheric Chemistry* 27:127–53.
- Beine, H. J., D. A. Jaffe, F. Stordal, M. Engardt, S. Solberg, N. Schmidbauer, and K. Holmén. 1997. "NO<sub>x</sub> during Ozone Depletion Events in the Arctic Troposphere at Ny-Ålesund, Svalbard." *Tellus, Series B: Chemical and Physical Meteorology* 49(5):556–65.
- Heintzenberg, Jost, Peter Tunved, Martí Galí, and Caroline Leck. 2017. "New Particle Formation in the Svalbard Region 2006 – 2015." *Atmospheric Chemistry and Physics* 17:6153–75.
- Hirdman, D., K. Aspö, J. F. Burkhart, S. Eckhardt, H. Sodemann, and A. Stohl. 2009. "Transport of Mercury in the Arctic Atmosphere: Evidence for a Springtime Net Sink and Summer-Time Source." *Geophysical Research Letters* 36(12):1–5.