

We would like to thank Reviewers 1 and 2 for the thorough revision of the manuscript and insightful comments that allowed to improve the quality of the paper and make more solid conclusions.

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 O₃ 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 NO₂/NO_x ratio and NO₂ produced through PAN decomposition, and long-term observations from Ny-Ålesund, weather regime and trajectory data are utilized to confirm weather regime 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.

A point-by-point response to the reviews is given below with Reviewers' comments shown in cursive and response shown in normal font.

1. Reply to the Reviewer 1

The paper is based on a novel set of NO_x and ozone measurements from three settlements on Svalbard during spring 2017. The main objective (page 4 lines 89-90) is to: identify specific factors affecting the concentration of measured compounds and define conditions that promote accumulation of local and long-range transported pollution in all three settlements..

The paper do address relevant scientific questions within the scope of ACP, and it does present novel data from the measurements on Svalbard and applies a semi-novel concept linking atmospheric circulation regimes and pollution levels.

Although several possible environmental impacts are mentioned in the introduction, objective/science questions on page 4 and the discussion/conclusion sections it becomes clear that the primary objective is to study the air quality (wrt NO_x and ozone) in the three settlements and how this is related to atmospheric conditions.

The main issue with the paper is the inconsistency of the data. This relates to the lack of co-location for the NO_x and ozone measurements, no ozone measurements in Adventdalen, and the issues with calibration of the instruments in Barentsburg (as also pointed out the authors at the end of the discussion section). This has effects on the conclusions that can be drawn, in that these can not be very strong. The manuscript includes in several places statements like "This may be explained by the fact that the measurement station in Ny-Ålesund was located much closer to the diesel power plant, ". Also, the discussion linking circulation regimes and air quality lacks clarity and could benefit from including more years (using ozone data from Zeppelin) to get a more robust result.

I find that the description and discussion in many places could have been more precise (cf detailed comments below).

I suggest that the manuscript can be published after major revisions.

Reply to the major comments

We agree that the lack of NO_x/O₃ instrument co-location in Adventdalen and Ny-Ålesund during the 2017 campaign is a major drawback of this study. The NO_x 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_x monitor was installed there due to the limited grant funding.

Regarding NO_x and O₃ 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_x and O₃ 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_x 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 24th of April to 13th 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 O₃ 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 O₃ observations at the Zeppelin station were not significantly affected by local NO_x 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_x pollution on the O₃ concentrations at the Zeppelin station have been modified accordingly (lines 288-305 in the new version of the manuscript).

To investigate the influence of local NO_x emissions on the O₃ 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_x emissions on the O₃ 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 O₃ sonde launches were available for the 2017 campaign (Figure 4). However, the long-term data below 100 m from the O₃ sonde profiles may be used to study influence of the local NO_x pollution in Ny-Ålesund on the O₃ concentration. These observations are suitable for this purpose because the O₃ sonde launching facility is located just 200 m to the south-south-west and 500 m to the south from the NO_x monitor and diesel power plant, respectively. Thus, when the monitor detected NO_x concentration above long-term springtime average in the launch hour, the influence of locally polluted air masses might have been observed in the lowest O₃ sonde data. There were in total 59 O₃ sonde launches, for which NO_x monitor data was available in spring 2009, 2010, 2015, 2016, 2017 and 2018. The O₃ profile data in the lowest 100 m have been extracted for all 59 launches and grouped according to the NO_x concentration detected by the monitor and wind direction in the O₃ sonde profiles: 1) above mean NO_x concentration and northerly wind direction; 2) below or equal to mean NO_x concentration and northerly wind direction. The median and mean O₃ values below 100 m in the group where the NO_x values were above NO_x mean were 11 % and 15% lower, respectively, than for the second group with northerly winds, but without elevated NO_x concentration. Thus, the O₃ concentration in lowest 100 m downwind from the power plant in the settlement may be reduced significantly due to local NO_x emissions, but the frequency of such events is unknown in absence of continuous O₃ measurements in the village.

Following the advice of the Reviewer 1, we included analysis of more years into the discussion part to get a more robust result linking weather regimes and air quality (lines 567-611).

To get a more robust result linking weather regimes and air quality, we would like to compare long-term springtime (23 March-15 May) weather regime data with NO_x data from Ny-Ålesund, O₃ concentration from the Zeppelin station, O₃ sonde and radiosonde data as well as FLEXPART trajectories for a period from 1990 to 2018. The FLEXPART and weather regime data were available for all years, while there were gaps in observational data from Ny-Ålesund. The data availability chart is shown in Figure A3 indicating number of hourly measurements for surface NO_x and O₃ data and number of radiosonde and ozone sonde launches per spring season each year. The hourly O₃ data is available for all years, while hourly NO_x data was only available in 2009, 2010, 2015, 2016, 2017 and 2018. After spring 2018, the NO_x monitor was moved to other location in Ny-Ålesund, therefore 2019 - 2022 data are not included in the current analysis to keep measurement consistency. The O₃ soundings and radiosonde AWI's datasets start in 1992 and 1993 with median number of radiosonde soundings and O₃ soundings per spring season being 54 and 11, respectively.

The box and whisker plots of O₃, NO_x and temperature inversion strength (TIS) for different weather regimes are shown in Figure A4. As during the 2017 campaign, the highest median O₃ values were detected for ScTr regime, while Zonal regime (ZO), during which O₃ values were also higher, was absent during the field work (Figure A4a)). The ZO regime is characterized by negative geopotential height anomaly at 500 hPa centred between Iceland and southern tip of Greenland and southerly flow over Svalbard (Papritz and Grams, 2018). The lowest median O₃ values in the long-term data are for GL regime and for European blocking (EUBL) that was absent in spring 2017. The variability of O₃ concentrations (range between 25th percentile and 75th percentile) for the EUBL regime is also remarkably higher than for other regimes. The EUBL regime is characterized by the positive 500 hPa anomaly centred over the North Atlantic. This promotes transport of air from south-west and west to Svalbard (Papritz and Grams, 2018). Long-term trajectories for the different regimes are shown in Figure A5. The lowest percentage of trajectories descending from higher altitude (>2000 m) is modelled for GL regime, while the highest percentage of elevated trajectories is obtained for EUBL, ScBL and ZO regimes. No specific trajectory probability pattern may be defined for "no regime" (Figure A5a), while distinct long-range transport signatures are identified for other seven regimes (Figure A5 b)-h)). In addition to the air transport path and trajectory altitudes, the sea-ice conditions and BrO concentration are important factors affecting the concentration of O₃ in each particular season.

Similar to the 2017 results, the highest median NO_x values are observed for "no" regime, ScTr, but two other regimes, when the long-term median NO_x values are high as well, EUBL and ZO, were not present during the campaign (Figure A4b)). The EUBL show pronounced transport of air masses from the west to Svalbard (Figure A5f). Thus, strong westerly component of the wind at the measurement stations and significant changes in local pollution dispersion conditions are expected for this regime as was observed during ScBL regime in 2017 when westerly component of the wind was present as well. Temperature inversions are common phenomena at high latitudes, in particular during the cold seasons due to radiative cooling of the surface and descending motion and heat advection from the south aloft. The inversions were detected in 27% of all the days in the measurement campaign period in 2017. This frequency of inversion occurrence is quite low in comparison with the results from previous studies of Dekhtyareva et al. (2018), where it was observed in 60% of the springtime profiles in 2009. Despite low frequency of occurrence, temperature inversions have significant influence on the dispersion efficiency, and, hence, according to the WRS-test, the median daytime (from 06UTC to 18UTC) concentrations of NO_x were higher ($p < 0.05$) at all three stations for the days when this phenomenon was observed in the radiosonde data in 2017. In the long-term data, the median temperature inversion strength was high for no regime, GL, AR and ScBL, but the highest median TIS was for Atlantic trough (AT) regime, the regime that was absent during the campaign (Figure A4c)). The AT regime is characterized

by a negative 500 hPa geopotential height anomaly to the east of Ireland and high cyclone frequency in that region (Papritz and Grams, 2018), while the cyclonic activity around Svalbard is lower, and these conditions may promote strengthening of the temperature inversion.

Thus, the results of the weather regime analysis performed for 2017 campaign are representative for characterization of the influence of different synoptic scale conditions on the NO_x and O₃ concentration in Ny-Ålesund. However, the three regimes that were absent during the 2017 campaign (AT, EUBL and ZO) are important in the long-term statistics for NO_x, O₃ and TIS in the settlement.

Reply to specific comments

Page 2, line 43. There is a very appropriate reference to Beine et al., 1997 who found that PAN decomposition was an important source of NO_x at the Zeppelin station. However, this point has not been taken up again during the discussions.

We would like to thank the Reviewer 1 for the useful comment. We have included the following part on PAN decomposition into the discussion (lines 501-522):

The results from radiosonde and ozone soundings, CO and particle measurements, presented in this study, demonstrate that the O₃ observations at the Zeppelin station were not sensitive to the local NO_x pollution from Ny-Ålesund, and thus were representative as background values for comparison with Barentsburg and investigation of the influence of prevailing long-range transport patterns on the measurements at these stations. Furthermore, O₃ data from the Zeppelin station may be used to assess how the PAN decomposition might have affected the background NO_x concentrations in Svalbard during the 2017 campaign.

Previous studies have shown that the NO_x/PAN ratio increases at temperatures above -10 °C, and PAN decomposition becomes a major source of background NO_x in Svalbard (Beine et al., 1997a; Beine and Krognnes, 2000). As the temperature at the Zeppelin station varied from -22.7 to 0.8 °C during the campaign, we would like to investigate the contribution of PAN decomposition to the background NO_x concentration in Svalbard. The PAN decomposition rate may be estimated using several approaches (Beine et al., 1997a), here we apply a linear relationship between O₃ and PAN concentration derived from previous measurements at the Zeppelin station: $PAN[ppt] = (O_3[ppb] - 26.58) / 0.034$ and then calculate PAN decomposition rate (Beine and Krognnes, 2000) (Figure A2a). The maximum PAN decomposition rate has been calculated using temperatures and O₃ concentration observed at the Zeppelin station applying equation (1) from Beine and Krognnes, 2000 (Beine and Krognnes, 2000). The depletion events when O₃ concentration was below 26.58 ppb have been excluded from the calculation (Beine and Krognnes, 2000). The median calculated PAN concentration of 356 pptv (ppb × 10⁻³) is comparable with previous springtime Arctic observations (Beine and Krognnes, 2000; Kramer, 2015). The estimated maximum PAN decomposition rate for the whole campaign varied from -0.0033 to -17.2 pptv hour⁻¹ with median value of -1.29 pptv hour⁻¹ (Figure A2b). The maximum PAN concentration coincides with the strongest O₃ increase event occurred 03.05.2017 (Figure 10b). The temperature increased simultaneously for that day (Figure 2a) promoting efficient PAN decomposition (Figure A2b). Applying Theil's non-parametric regression with the slope of -5.07 (pptv NO_x/pptv hour⁻¹ PAN) suggested by Beine et al., (1997a) for Svalbard, the background concentration of NO_x would be 87.2 pptv. However, these concentrations are too low for the equipment used in the 2017 campaign to detect the variations in the concentrations caused by the PAN decomposition.

Page 3 line 68: Is it really true that there is no diurnal cycle in the demand for electricity in Longyearbyen? What about the use for cooking and light with a peak in the afternoon?

We would like to thank reviewer for the comment. The sentences have been corrected as following (line 68-71):

In contrast to the energy needed for heating, the energy demand for electricity production is mostly independent on the air temperature. Industry, business and communal buildings stand for more than 70% of the electricity consumption in Longyearbyen. There is a diurnal variation in the power demand with higher daytime values in winter.

Page 7 lines 160-170. It is unclear what this “no regime” is. The “for example” wording on line 160 is very confusing.

The sentence has been changed as following (lines 180-182):

In the climatological mean, the large-scale conditions are characterized by weak ridging of absolute geopotential height at 500 hPa over the eastern North Atlantic and westerly upper level flow over Svalbard. Such regime is placed in the "no regime" category in the Grams et al., 2017 classification (their Fig. S1h).

Page 7 lines 167-171. The sentence starts with “Secondly”. Does this means that the identification of the flow regimes for the sub-periods was done on another dataset than the ERA5?

Following sentence has been added into the manuscript: (line 172).

In the current work, we apply Dr. Christian Grams’s weather regime classification that is based on the 6-hourly ERA-Interim data.

Page 10 line 248: The amplitudes are not very high (+/- 2ppb or so), but they are very short term fluctuations.

The sentence has been corrected as following (lines 277-278):

The Barentsburg O₃ data contains some abrupt peaks with magnitude of up to 9 ppbv and duration of just one hour (light blue line in Figure 3c)), while they are absent in the Zeppelin O₃ data.

Page 11 line 268: Define ozone titration efficiency.

The sentence has been change as following (lines 306-308):

Difference between the original and smoothed O₃ data from Barentsburg varies from -19% to 11% of the smoothed value, and there is a moderate negative correlation between the magnitude of NO_x peak and reduction in O₃ concentration ($r=-0.65$, $p<0.0001$).

Page 11 line 267-271: First, it is stated that there is a strong negative correlation, and then later (line 270) it is stated that it is not statistically significant. This seems contradictory.

The sentence has been rewritten as following (lines 308-311):

Despite this sensitivity of O₃ concentration to local pollution in Barentsburg, the median NO_x concentrations observed there were low and average reduction of O₃ concentration in comparison to the smoothed values was only 1%. This effect is not statistically significant, and therefore other factors, such as variation in concentrations within long-range transported air masses, masses, may be more important for explanation of difference between the O₃ Zeppelin and Barentsburg datasets.

Page 11 line 281. The word “However” seems misplaced here. Since the measurements are so close to the source this kind of extreme values can be expected there.

The word “However” has been deleted.

Page 10 line 247. *It is concluded that the synoptic conditions have minor effects on NO_x due to the low correlation between Ny-Ålesund and Barentsburg. However, from the maps (figure 6) it is clear that in Ny-Ålesund the source is North of the station, while it is the opposite in Barentsburg. Thus, I would expect that the wind direction component of the synoptic conditions could give negative correlation.*

We would like to thank the Reviewer for the comment. Indeed, one may expect negative correlation for the stations, but the atmospheric stability also plays role, since the monitoring station in Barentsburg is located on the hill above the pollution source, while stations in Ny-Ålesund and Adventdalen are located on the same level or below emission sources. The combination of these factors caused by the synoptic situation affect the correlation of the measurement results from the three stations. The sentence has been changed accordingly (lines 271-276):

On the contrary, no correlation is present with NO_x data from Barentsburg. Low correlation between the NO_x 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_x 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_x values due to long-range transport to Svalbard would be undetected in the NO_x datasets presented in the current study.

The paper seems to neglect the effect of reaction R2 for the NO₂/NO_x ratio. With appropriate values for k1, [O₃] and k2 (the photolysis rate) one can derive the steady state NO₂/NO_x ratio. It would be of interest to see how the ratios observed in Adventdalen during daytime is affected by the photolysis.

We would like to thank for this comment. The section about the effect of photolysis is added in the discussion (lines 474-501):

The NO_x monitor in Adventdalen was located far away from stationary emission sources and showed the highest daytime NO₂/NO_x ratio (Table 1). We would like to investigate how the ratios observed there were affected by photolysis. The photolysis rate of NO₂ depends on solar zenith angle (Parrish et al. , 1983), which in turn depends on day of year. Measurements were performed between days 81 and 134, and the noon solar zenith angle in Longyearbyen area varied from approximately 77° to 62° (Robertson et al. , 2006). Following equation (15) in Parrish et al. (1983), minimum clear-sky photolysis rate for the start of the campaign was 0.0026 s⁻¹, and maximum clear-sky photolysis rate for the end of the campaign was 0.0061 s⁻¹ (black squares in Figure A1a)). There are many factors that affect NO₂ photolysis rate, such as aerosol load, clouds, water vapour content and surface albedo (Trebs et al. , 2009). The albedo may significantly increase the NO₂ photolysis rate (Trebs et al. , 2009), and Dickerson et al. (1982) suggested albedo of snow with respect to j(NO₂) to be 93%. Trebs et al. (2009) suggested in their equation (2) a polynomial fit between global irradiance and NO₂ photolysis rate that includes both clear-sky and cloudy conditions and takes into account the contribution of albedo. The albedo calculated as the ratio of upward and downward short-wave radiation measured by CNR1 Kipp Zonen net radiometer in Adventdalen and observed global radiation were used to estimate j(NO₂) (red line in Figure A1a)). Figure A1b) shows NO/NO₂ ratio calculated using O₃ concentration measured in Barentsburg (closest station where O₃ measurements were available), j(NO₂) and temperature-dependent rate coefficient kNO+O₃ obtained using temperatures in Adventdalen (equation 6.6 and Table 6.1 in Seinfeld Pandis, 2006). The peaks of NO/NO₂ ratios are especially pronounced for the days with decreased O₃ concentration (01.04.2017 and the period 485 from 04.05.2017 to 09.05.2017). Note, the calculation is based on the O₃ data from Barentsburg, thus this introduces an uncertainty in the exact NO/NO₂ ratios estimated for Adventdalen. The observed and calculated NO₂/NO_x ratio for Advendalen are shown in Figure A1c). The missing data in the observed NO₂/NO_x ratio (blue line) indicate that

both NO and NO₂ values were within zero-noise level, while missing data in the calculated NO₂/NO_x ratio is due to missing O₃ observations in Barentsburg. The observed and calculated values are of the same order, but NO₂/NO_x ratio is underestimated in 64% of all available data, especially for the days with low O₃ values. This underestimation was present even in hours influenced by fresh local NO emission (green line) and might have resulted from the modelling errors that could occur if the surface albedo was high (Trebs et al. , 2009) or because the actual O₃ values in Adventdalen were lower than in Barentsburg. The NO₂/NO_x ratio is overestimated in 31% of all available data. In these hours, the actual O₃ concentration might have been higher in Adventdalen than in Barentsburg (used for calculations). The most pronounced overestimation is noticeable in the period from 26.04 to 29.04 when NO values in Barentsburg were higher than in Adventdalen, and thus more pronounced O₃ titration with local NO might have occurred in Barentsburg.

Figure 4. The diurnal cycle of the means of ozone at BB and Zeppelin are shown. These are the means over the springtime period (about 55 days) I presume. Please add to this figure the standard error of the mean for each hour, so that we can see if these diurnal cycles are really statistically significant.

The standard error of the mean has been included for each hour (Figure 5 in the new manuscript). To improve readability of the figure, separate subplots a) and b) have been made for NO_x and O₃ data, and supporting text has been modified accordingly.

Page 12, line 305. I don't understand the argument that enhanced photolysis is compensated by convection. Photolysis in itself would not reduce ozone significantly as the reaction $O + O_2 + M \rightarrow O_3 + M$ would very rapidly reform ozone. In addition, I would expect that for Zeppelin convection would mix in PBL air with lower ozone. At very low NO_x levels there could be enhanced loss of ozone during daytime through $O_3 + OH \rightarrow O_2 + HO_2$ followed by $O_3 + HO_2 \rightarrow OH + 2 O_2$

Page 12, line 307: If the diurnal cycle is not statistically significant, there is no need to discuss possible physical/chemical explanations!

The sentences have been modified as following (lines 359-364):

In contrast, the Zeppelin station is located at the altitude of 474 m a.s.l. and mostly samples air from the free troposphere with higher O₃ 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 O₃ concentrations measured at the stations (Table 1).

Page 12 line 312: Wind speed 4.1 m/s. This must be the average wind speed. Please also give the variance.

The standard deviation of the mean has been included in the sentence (line 365).

Page 13 line 317: You have written: normally ventilation is sufficient to remove NO_x emitted by the usual amount of motorized traffic. I don't understand this statement: is NO_x completely removed?

The sentence has been modified as following (lines 375-378):

Such low wind speed is untypical for the wind regime in Adventdalen, where normally ventilation is sufficient to effectively disperse NO_x emitted by the usual amount of motorized traffic.

Page 15 line 342: Unclear sentence. Is the 46% referring to the whole period or sub-period I? Is 0.95 °C the median or is it the deviation from the median during sub-period I.

The sentence has been corrected in the following way (lines 401-402):

In the sub-period I, the temperature inversions were observed in 46% of the radiosonde profiles from Ny-Ålesund, but the median inversion strength was below 0.95°C (median for the whole campaign).

Figure 7. It would be useful to have the sub-periods indicated over the individual plots. The figure has been modified as suggested by the Reviewer (Figure 8 in the new manuscript).

Page 14, Section 3.2: I find this whole section quite unorganized. The whole section seems to focus on PBL high and how it affects NO_x , and possible transport patterns for ozone that allow ozone depletion events. There is a lack of motivation for selecting these regimes. E.g. the regimes depicted in fig 7a and 7h looks very similar to me, and without a rationale for splitting this in two different regimes. A factor that is completely missing is the possibility of tropopause folding events with intrusion of ozone rich air, presumably related to the circulation regimes.

We would like to thank the Reviewer for the comment. The weather regimes were identified as described in Section 2.4 based on the prevailing large-scale atmospheric circulation patterns and 500 hPa geopotential height. This classification was done as described in Grams et al., 2017, independently on the in-situ meteorological or chemical data. The same large-scale regimes occurred in different sub-periods throughout the campaign were analysed separately because the sea-ice conditions, snow cover, insolation, temperatures and boundary layer height evolve as seasonal transition takes place from March to May in Svalbard. Thus, even if large scale transport patterns may look similar, the concentrations of measured compounds may be different due to local processes.

The downward transport from the free troposphere is, indeed, a significant source of lower-altitude O_3 in Svalbard region, particularly during winter and spring (Hirdman et al., 2009). Thus, not only low-level trajectories need to be taken into account, but also percentage of trajectories descending from upper levels (>2000 m) during different sub-periods.

The respective discussion the height of transported air masses for different periods obtained in FLEXPART and HYSPLIT trajectories and importance of trajectory height for the O_3 concentration has been included in lines 451-459:

The O_3 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). As in previous studies of Hirdman et al. (2009), the downward transport of O_3 -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_3 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_3 concentrations in different sub-periods (e.g. ScTr regime in Figure 9c) and e) and Table2).

On the transport of pollutants to Svalbard the work by Hirdman et al. should be referenced (Hirdman et al., Atmos. Chem. Phys., 10, 9351–9368, 2010 www.atmos-chem-phys.net/10/9351/2010/ doi:10.5194/acp-10-9351-2010)

The reference has been included in the lines 192-193 as well as two additional relevant references on FLEXPART and Arctic long-range transported pollution (Hirdman et al., 2009; Hirdman, Burkhardt, et al., 2010; Hirdman, Sodemann, et al., 2010).

Page 16, line 366-380. Elevated NO_x concentrations were found in Adventdalen but not in Barentsburg for period VI with cold conditions and low PBL height. These conditions with low wind (and maybe clear sky) would I believe enhance recreational snow mobile traffic and thus emissions, which is much more pronounced in Longyearbyen than in Barentsburg. In general, there may be a link between weather and emissions that is not mentioned in the paper.

The observations of number of snowmobiles during the campaign were scarce, but the following might be stated (lines 367-373):

To test if the size of snowmobile motorcade has an influence on the NO_x concentration in Adventalen, manual observations of number of snowmobiles were done in 19 days. In general, the effect of large number of snowmobiles was noticeable in the NO_x data only in case of low wind speed. For example, in the evening of 01.05.2017, the wind speed was 1.9 ms⁻¹, and the NO₂ concentration increased sharply to 7.3 ppb due to 21 snowmobiles passing by the station. The group of similar size was passing by in the evening of 02.05.2017, but the effect on NO₂ values was three times lower as the wind speed was higher (4.0 ms⁻¹).

Page 18, line 382-390. The paper concludes (elsewhere) that there is high correlation between ozone at Zeppelin and Barentsburg, thus the measurements represent region ozone levels. Since ozone data from Zeppelin is available for a number of years, this regime analysis could be extended for a much longer period and thus be much more robust.

The long-term regime and Zeppelin O₃ data analysis has been included in the discussion part of the paper (lines 567-611).

Figure 8:

-This is not really the trajectory probability for the different regimes, but rather for the sub-periods. Having a longer (multi year) record to make these probabilities for the regimes would help. Very difficult to read the red contours.

- The maps are very small. There is no need to include the same label bar 9 times. Also I recommend that each map is labeled with the name of the corresponding circulation regime (applies also to figure 7).

The figure (Figure 9 in the new manuscript) has been modified as suggested by the Reviewer 1.

The long-term trajectory probability for different regimes have been included into the Discussion part of the paper (lines 566-610).

Page 19, line 396. Why is the HYSPLIT model used for these back trajectories and not Flexpart?

The FLEXPART dataset used in this study is a long-term global dataset, and trajectories were not run from a specific point, while the online version of HYSPLIT allows to easily set up a starting location and height of Zeppelin and Barentsburg stations and run a trajectory simulation for the extreme case studies discussed in the paper. Similarly, the combination of HYSPLIT, Lagranto and FLEXPART has been used in Trickl et al., 2020 to study stratospheric intrusion events.

Page 22, line 474. The authors claim that “The weather regime approach is novel in the air pollution research”. However, this has been used in several studies before, although not for the Svalbard region I believe.

We would like to thank the Reviewer for this comment. The respective references have been included in the methods part (lines 165-171) in addition to another relevant article about connection of NAO and air pollution transport (Eckhardt et al., 2003; Ibrahim et al., 2021; Ménégos et al., 2010), and line 626 has been corrected as following:

The weather regime approach is novel in Svalbard air pollution research.

2. Reply to the Reviewer 2

Reply to the major comments

The observations itself are of interest because in the Svalbard are really sparse, so they can help to understand formation and evolution of O₃ in a remote area effected by interesting and mixed effects such us depletion due to reactive halogen compounds and photochemistry due to local well defined emission sources. However, there are several shortcomings that limit a lot the results of this paper, therefore I'm really skeptical to suggest to accept this manuscript for publication on ACP, unless it will be deeply revised, for the following reasons:

1) Only in the Barentsburg Station are observed both O₃ and NO_x, that are the two species fundamental for this study, in the Adventdalen and Ny-Alesund sites are missed the measure of O₃, therefore a comparison and correlations of these species among these station is misleading. To partially overcome this problem for the Ny-Alesund analysis are used the O₃ measurements of the Zeppelin observatory, but this is not the solution not only because the latter is 2 km away from Ny-Alesund site, but, more seriously, it is on the top of a mountain at 474m. a.s.l., whereas Ny-Alesund site is at 23 m. a.s.l. and near the sea. Finally, since in Ny-Alesund site are missed also meteorological measurements, for the analysis on this site are used data of the top of the mountain (Zeppelin station). It would have been much more worth to install the NO_x analyser in the Zeppelin station where meteorological data and O₃ measurements were available then in the Ny-Alesund site.

We agree that the lack of NO_x/O₃ instrument co-location in Adventdalen and Ny-Ålesund during the 2017 campaign is a major drawback of this study. The NO_x 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_x 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_x data from the village (p. 5 line 120 in the manuscript). The O₃ and CO data from the Zeppelin station have been combined with the meteorological data from the Zeppelin station.

Regarding NO_x and O₃ 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_x and O₃ 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_x 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 24th of April to 13th 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 O₃ 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 O₃ observations at the Zeppelin station were not significantly affected by local NO_x 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_x pollution on the O₃ concentrations at the Zeppelin station have been modified accordingly (lines 288-305 in the new version of the manuscript).

2) Lines 233-236: Since the CO measurements were stable at Zeppelin station so no sharp peaks to identify local emission were detected, this is a proof that the measurements at Ny-Alesund are not useful to understand potential impact of local pollution on O₃ evolution in the mountain top.

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_x pollution on the O₃ concentrations at the Zeppelin station have been modified accordingly (lines 288-305 in the new version of the manuscript).

3) Lines 243-247: Here the Author affirm that local emissions are important because the correlation between NO_x measurements at Ny-Alesund and Adventdalen sites are weak, but at lines 233-236, looking at CO data they assert that local pollution are not important for the area under investigation. This is contradictory conclusion is due to another big issue: since CO measurements are available only at Zeppelin station, the signature of local emissions in other sites (where CO were not measured), were tried to find in the correlation between NO_x observation, again measurements of CO in Ny-Alesund and Adventdalen site would have been worth to make this conclusion.

The sentences have been modified as following (lines 271-276 in the new manuscript):
Low correlation between the NO_x 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_x 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_x values due to long-range transport to Svalbard would be undetected in the NO_x datasets presented in the current study.

4) Lines 255-257: Author here assert that synoptic transport is more important than local emission looking, now, at the correlation of O₃ measurements at Zeppelin and Barentsburg site, a couple of issues: a) $r = 0.69$ is considered a 'strong correlation', it means $r^2 = 0.47$, that is not that 'strong', b) again since O₃ is missed in the Ny-Alesund and Adventdalen, now to decide if dominate local emission or synoptic transport the correlation of O₃ between Zeppelin and Barentsburg site are used, while before (lines 233-236) were used NO_x for Ny-Alesund and Adventdalen sites, obtaining contradictory results.

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) Lines 295-298 and figure 4: The O₃ diurnal cycle of Barentsburg site is, as expected, completely different of that of the Zeppelin station, in the first is evident the typical profile dominated by photochemistry, in the later, a typical mountain station data, with no diurnal cycle. This is what expected, but again in contrast with what reported in lines 255-257 where Author affirm that the O₃ measured in these two sites showed a 'strong correlation': they have a completely different

dynamics, as can be expected since one is in a mountain and the other in a site at 40 m. a.s.l.

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_x and O₃ 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 O₃ 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 O₃ concentrations measured at the stations (Table 1).

6) Lines 385-399: Here initially, looking at trajectory analysis Authors affirm that the O3 decrease can be explained by local depletion due to air masses rich of BrO, whereas at the end is supposed that may be due to less photochemistry due to 'lack of sunlight and O3 precursors such as NOx'. A good result of this analysis would have been if the two effects (depletion due to BrO vs photochemistry and NOx emission) were well characterized and, from observations and model analysis, quantified and compared, unfortunately here both effects are claimed, as can be guessed even without any kind of measurements and/or model analysis.

Following recommendations of the Reviewer 1, we included investigation of possible downward transport of O₃ 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 O₃-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 O₃ 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 O₃ concentrations in different sub-periods (e.g. ScTr regime in Figure 8c) and e) and Table2). (lines 452-455).

The discussion about NO_x transport and PAN decomposition modelling has been included in the Discussion part of the paper (lines 501-522).

7) Lines 408-411: Finally, the Authors, looking O3 sondes data confirm that Zepellin data are different of that in Barentsburg because one is at the top of Mountain and the other at 40 m. a.s.l. I think that this would have been the first analysis of the paper, and not the last one after different correlation analysis where was mentioned that O3 data of those sites are strongly correlated.

Indeed, there is a difference between the O₃ 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) *Lines 471-473: From the data and analysis I'm not comfortable with the conclusion that local emission of NO_x may reduce O₃ level by few percent in the Ny-Alensund site, since O₃ there is not measured, but, again, are used for this conclusion O₃ measured at the top of mountain. Here for example, since O₃ where not measured would have been worth to use a Box model (such as MCM) to model O₃ at Ny-Alensund, constrained by local NO_x measurements.*

8) To investigate the influence of local NO_x emissions on the O₃ 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_x emissions on the O₃ 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 O₃ sonde launches were available for the 2017 campaign (Figure 4). However, the long-term data below 100 m from the O₃ sonde profiles may be used to study influence of the local NO_x pollution in Ny-Ålesund on the O₃ concentration. These observations are suitable for this purpose because the O₃ sonde launching facility is located just 200 m to the south-south-west and 500 m to the south from the NO_x monitor and diesel power plant, respectively. Thus, when the monitor detected NO_x concentration above long-term springtime average in the launch hour, the influence of locally polluted air masses might have been observed in the lowest O₃ sonde data. There were in total 59 O₃ sonde launches, for which NO_x monitor data was available in spring 2009, 2010, 2015, 2016, 2017 and 2018. The O₃ profile data in the lowest 100 m have been extracted for all 59 launches and grouped according to the NO_x concentration detected by the monitor and wind direction in the O₃ sonde profiles: 1) above mean NO_x concentration and northerly wind direction; 2) below or equal to mean NO_x concentration and northerly wind direction. The median and mean O₃ values below 100 m in the group where the NO_x values were above NO_x mean were 11 % and 15% lower, respectively, than for the second group with northerly winds, but without elevated NO_x concentration. Thus, the O₃ concentration in lowest 100 m downwind from the power plant in the settlement may be reduced significantly due to local NO_x emissions, but the frequency of such events is unknown in absence of continuous O₃ measurements in the village.

9) *Lines: 484-486: From data analysis and model simulation it is hard to support these conclusions.*

The sentence has been modified as following (lines 636-667):

In contrast to NO_x, the concentrations of O₃ 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 O₃. In other words, both these stations are regionally representative for the O₃ concentrations.

A general comment and a suggestion for further observations in this area: NO₂ measurements in remote area, where the concentrations are very low may have bias or instruments are below the detection limits for most of the times. There are several papers that suggest to use instrument that measure directly NO₂, using CAPS, LIF or CRDS techniques (actually CAPS are commercially available now) or, at least, chemiluminescence systems that uses photolytic conversion of NO₂ into NO, besides systems like those used in this work (model T200) that uses molybdenum oxide converters (Steinbacher et al., 2007; Dunlea, et al. 2007; Yang et al., 2004; Villena et al., 2012).

We would like to thank Reviewer 2 for the suggestion of the alternative NO_x 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_x data from Ny-Ålesund that is included in the new version of the manuscript.

We will definitely consider suggested measurement techniques for background NO_x observations during planning of new field campaigns in the Arctic.

Reply to minor comment

Line 391: When in a time period, 67% of data are missed analysis and conclusion are very weak, so may be better not include that period in the analysis.

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 O₃ 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).

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