Response to Anonymous Referee #1 Comments

We thank this reviewer for the constructive feedback on our manuscript. Our responses and changes implemented in the manuscript are summarized below in blue bold font after each of the reviewer's comment points.

GENERAL COMMENTS

This study reports observations of air and snow chemistry from a 14 month measurement campaign at Dome C, East Antarctica with a focus on pollution episodes caused by the year-round operated Concordia station. Gasphase concentrations of nitrogen oxides (NOx) increased by several orders of magnitude during a total of 15 events of pollution transport from the station during the study period, with concurrent decreases in ozone indicating titration by nitric oxide. While near-surface air concentrations of NOx settled to background levels within hours, firn air concentrations remained elevated for days to up to a week. The snow chemical composition may be affected as indicated by nitrate concentrations in snow below the top layer being higher near the station than further away. The authors conclude that these observations provide insight to the discussion on how representative snow photo-chemistry studies near research stations are for the wider Antarctic ice sheet.

The detection of elevated NOx and associated O3 decrease when sampling station power generator exhaust, and of course also any kind of motorised traffic (air plane, ski doo, tractors etc) is not new, but a well-known limitation of studies of background atmospheric chemistry at most polar research stations. This is typically mitigated by careful filtering of atmospheric measurements.

A new insight from this study is the observation that pollution plumes diffuse into the upper snowpack and remain there for several days, with the potential to alter chemical composition of the upper snowpack near the station and therefore compromise local snow photochemistry being representative for the wider ice sheet. Understanding changes in air and snowpack chemistry near Concordia station would be potentially very useful for the design of future experiments at Dome C and elsewhere. Rather than just casting doubt on previous measurement campaigns I suggest the authors improve the manuscript by broadening the range of considered chemical species and by providing more quantitative detail in their discussion of pollution impacts on snow chemistry as promised by the title:

The abstract states that other reactive chemical species were also measured, notably formaldehyde and gaseous elemental mercury (GEM). Even though not as sensitive to pollution as NOx I urge the authors to present and discuss those data during pollution events to achieve a more general picture of pollution impacts near the station. For example formaldehyde has been discussed as a snowpack HOx radical precursor at Dome C (Preunkert et al., 2015), and it would be important to understand if and how station pollution changes also nearby CH2O profiles in snow.

We re-examined the available formaldehyde and GEM data from the campaign. Those measurements do not quite have the time resolution and sensitivity as the NOx and ozone monitoring. Enhancements in ambient samples suspected to be influenced by station emissions have previously been noted by in measurements taken during the OPALE campaign in 2011

[*Preunkert et al., 2015*]. Formaldehyde measurements overlapped with the ozone and NOx monitoring only for a short period during the 2012/2013 austral summer, and this measurement period suffered from a number of instrument problems. The remaining data did not allow a conclusive evaluation on the behavior of formaldehyde in the snowpack during pollution events. We also revisited the gaseous elemental mercury (GEM) measurements taken during the campaign [*Angot et al., 2016*]. These data did not show a clear signature of GEM in snowpack air that correlated with the NOx enhancements during pollution events. This information has been added to the manuscript text. There are no other chemical gas measurements available from the campaign for a further investigation.

Snow nitrate profiles observed in the clean-air sector near the station show a less steep decline with depth and slightly higher concentrations than at sites further away (>5km). Please discuss the role of spatial variability in snow accumulation and stratigraphy. The presence of sastrugi as well as substantial snow drift around buildings/obstacles as seen on Fig.1 certainly have an impact on snow chemistry profiles. Accumulation rate determines exposure time of a given snow layer to actinic flux as well as stratigraphy/ snow optical properties.

Authors' response: The skin layer of the surface snow has been collected at Concordia every 3 days for more than 8 years. Furthermore, every three months surface snow is collected randomly in an area of ca. 100 m^2 at the same time. The standard deviation in the nitrate concentration is ca. 20-30%. Manuscript Figure 8, and the new Supplement Figure 3 provide the available data for evaluation of the variability in nitrate results in snow samples collected from snow pits at different locations.

Can the authors provide an estimate of at least the order of magnitude of how much snow nitrate could be produced from pollution episodes throughout a year, knowing O3 and NOx in firn air & assuming HOx levels in summer?

Authors' response: We agree that this is an interesting and compelling question, but we do not see a way to possibly develop a nitrate production estimate given the available data and analysis tools.

I'd suggest to double-check with co-author Savarino, who has measured the stable nitrogen isotope 15N in atmospheric and snow nitrate over several years now at Dome C. Assuming d15N in NOx from the station plume is different from other sources, did they ever detect any evidence from existing measurements of d15N in nitrate in snow (or in atmospheric particulate no3) that would indicate a contribution from local pollution?

Authors' response: Thus far there has been no indication of 15N signals from plume pollution. It needs to be realized that that the amount of nitrate in the snowpack is orders of magnitude larger than the sum of total oxidized nitrogen gases in the atmospheric boundary layer. Therefore, influences on the differences in the isotopic signature of atmospheric nitrogen would be very difficult to detect in snowpack samples.

SPECIFIC COMMENTS

Abstract & l162: in the text and Fig.4 a total of 50 pollution events are reported, whereas the abstract states 15 occasions of plume transport, how do the different numbers arise? Please explain.

Authors' response: In total there were about 50 pollution events. Of those, we counted 15 events in total when there was a spike in the above surface NOx measurements followed by an increase of at least 1 ppb of NOx within the snowpack. This detail has been added to the text.

l96-115 Please clarify the schedule of the firn air sampling. Apparently inlets were switched between the 2 snow towers every 24 hr (caption of Fig.6)?

Authors' response: Yes, the reviewer is correct. We added this information to the method description as well.

l235-239 This section requires further clarification: e-folding depth depends in a non- linear fashion on both, absorbing impurities (BC, HULIS etc) as well as on snow density & grain size/shape (scattering). Thus, the variability observed (Warren et al., 2006; France et al., 2011) is not only due to BC deposition but in part also due to the snow stratigraphy present at DC.

Authors' response: The sentence was reworded to: "Contamination of the snowpack around Concordia station has been noted in previous investigations. [*Warren et al.*, 2006] reported an > 3-fold increase in snowpack black carbon concentration after the station was established in 2003, with black carbon levels in pre-2003 snow (from deeper depths) also being in closer agreement with collected snow from further distance to the station. Black carbon is one of the contributing factors for a decrease in light penetration into the snowpack [*Warren et al.*, 2006; *France et al.*, 2011; *Libois et al.*, 2013]. Consequently, the black carbon snowpack contamination results in a shallower e-folding depth compared to pristine, uncontaminated snow.

Fig.1: To better illustrate the location of the Atmospheric Observatory add a wind rose and boundaries of the clean air sector.

Authors' response: A wind rose and lines that indicate the border of the clean air sector were added to the figure. The revised figure is shown below.

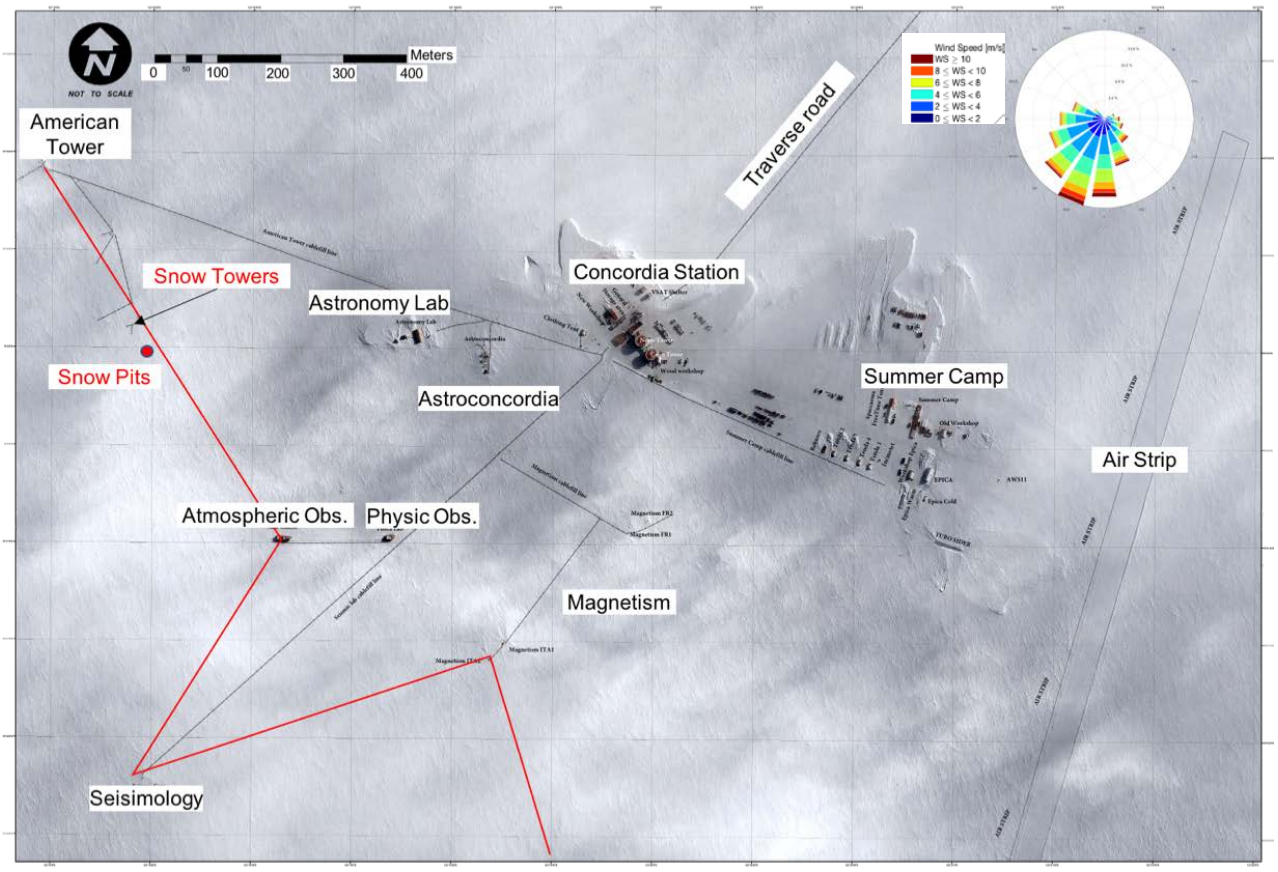

Figure 1: Satellite image of research station Concordia with location of the snow photochemistry experiment indicated by the red text. Its location was ~1 km west of the station main buildings and power generation plant. The map also indicates the location where the snow pits were collected, the border of the clean air sector (which is west of the red line), and a wind rose for a full year of wind data collected from the met tower that was adjacent to the snow tower manifolds.

Fig.2: Text only is hard to follow so please add a schematic indicating location and height/depth of all inlets and sensors, as well as of the snow pit profile.

Authors' response: We added this text to the figure caption: "(see Figure 1 in *van Dam et al.* (2015) for a schematic of a similar installation at Summit, Greenland)". Indication of the location of the snow pit sampling was added to Figure 1, and the snow pit profile data are shown in Figure 9.

Fig.6: caption "... measurements FROM the snow tower.." Supplement-Fig S-1 and S-2: in the online version no supplement was available. I suggest to show the O3 mixing ratios and the surface snow no3 also in the main manuscript.

Authors' response: The ozone graph was moved from the Supplement to the main manuscript. But we prefer keeping the surface nitrate graph in the Supplement as these data are purely ancillary supporting information.

REFERENCES

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