

We thank the reviewer for his/her comments that have improved the completeness and clarity of this manuscript. The comments by the reviewer are given below in black, our responses are marked in red and the corresponding changes to the manuscript are in *italics*.

## Interactive comment on “Characterization of Transport Regimes and the Polar Dome during Arctic Spring and Summer using in-situ Aircraft Measurements” by Heiko Bozem et al.

### Anonymous Referee #1

Received and published: 6 March 2019

This manuscript provides an analysis of air masses present during the 2014 and 2015 NETCARE airborne measurements spanning a broad region of the western Arctic between Spitsbergen and Alaska. An overview of the meteorological conditions during the summer (2014) and winter/spring (2015) campaigns is given, then trajectories are used to identify airmass history. Trace gas observations are described, then combined with potential temperature to identify regions with sharp gradients in CO and CO<sub>2</sub>. These gradients are used to define the "polar dome", the region of the cold, stable, near-surface Arctic airmass that is most isolated from midlatitude influences. (As this airmass wobbles over sources of pollution in the winter, it accumulates pollutants because sinks are very slow, leading to the seasonal near-surface "Arctic haze" phenomenon, which is of broad interest.) The statistics of CO and CO<sub>2</sub> abundance in the different regions identified from this analysis are then presented. Next, a "transport regime" analysis, based on the trajectories and using methods developed in Binder (2017), is used to evaluate the influence of lifting within and outside the Arctic, and diabatic processes, on CO and CO<sub>2</sub> mixing ratios within and outside of the polar dome. Finally, a discussion section evaluates the abundance of nucleation and Aitken mode particles within the three regimes identified in the earlier analysis (inside the polar dome, outside the polar dome, and a mixing region).

This is an ambitious manuscript, with many parts. It has interesting sections, but it doesn't seem to have a strong overall purpose. My fundamental complaint with the manuscript is that it doesn't make the case for any generality to the analysis. Are the results more broadly applicable outside of the narrow time period and location of the NETCARE airborne observations in 2014 and 2015? For example, in Sect. 5.3 there is a long discussion of how the CO and CO<sub>2</sub> observations can be used to identify the polar dome boundaries, and a specific range of potential temperatures and latitudes is the result of the analysis. This is great for these NETCARE observations, but are these findings more broadly applicable? For example, could one take the long-term surface observations at UtqiaĀvik (Barrow) or Alert or Zeppelin, apply the potential temperature and CO/CO<sub>2</sub> screens developed in this manuscript, and separate the data out into "in the polar dome" and "out of the polar dome" datasets? This would be useful to the scientific community. Without such broader relevance, this analysis is of interest only to the very small set of scientists interested in the NETCARE data.

We appreciate the reviewer's comments and the valuable suggestions for restructuring the manuscript which we largely followed and extended the discussion of the transport regimes. Regarding the point of a larger scale applicability of the method we included a first comparison with station data from Zeppelin in the reply.

In this particular paper our intension was indeed to develop an empirical dome boundary based on the airborne measurements of tracer like CO and CO<sub>2</sub> for NETCARE and to show first applications. We will extend the method and apply it to other airborne data sets or ground based data. Note however, that ground based data could eventually have a different tracer characteristics since these data are stronger affected by local sources and sinks and partly decoupled from the free troposphere by the boundary layer inversion. Nonetheless, we tested the approach for Zeppelin, which provides very promising results (see below). We will publish a follow-up paper, which will apply the method to a larger data set to investigate the general applicability.

In addition to my concern with the applicability of the findings, I feel the manuscript also needs restructuring. The paper opens with the meteorological analysis, which is fine. Next, though, is the trajectory analysis. It would be more logical if the next section were the identification of the polar dome using the trace gas gradients. Then the backtrajectories could follow, with backtrajectories initiated either within the polar dome, outside of it, or in the mixing region. The trajectories would then provide an independent and intuitive confirmation of the identification of the polar dome that was derived from the trace gas and potential temperature data.

We restructured the manuscript mostly as suggested: We moved the detailed trajectory analysis of former Figure 4 and 5 to the supplement and combined the chemical regime discussion based on trace gases with the (newly restructured) aerosol paragraph at the end. This results in a structure as suggested: Meteorological analysis, determination of the polar dome boundary, transport regimes and air mass history, chemical regimes. Note that Willis et al. (2019) and Schulz et al. (2019) already used the polar dome boundary presented in this study to analyse aerosol composition, transport and sources within the springtime polar dome and within and partly outside the summertime polar dome, respectively. For a more detailed aerosol analysis we refer to their studies and only briefly address characteristics of aerosol within this study.

Section 5.4, which is a presentation of PDFs of CO and CO<sub>2</sub> from the three different air masses is not very logical. CO and CO<sub>2</sub> were used to identify the three air mass classifications, after all, so it's entirely expected that they would have different PDFs—the reasoning is circular.

It is important to keep in mind that the dome boundaries were determined based on the gradients of CO and CO<sub>2</sub>, not applying any threshold value for tracers. In the case of horizontal (i.e. isentropic) gradients the analysis was performed for different isentropes in intervals of 2K, allowing for different thresholds for each isentropic interval. There is no step within the analysis, which makes use of an absolute value of either CO or CO<sub>2</sub>. Since the gradient is not related to an absolute value of CO or CO<sub>2</sub>, trace gas values and particular the variability therefore still provide additional information for the regimes. The width of the distributions should be larger outside the dome regions, the mean (median values) resemble the surface network data. We therefore kept this section since it illustrates the chemical properties of the regimes. As mentioned before it is moved to the end of the manuscript.

Instead of this section, the next logical section would be examining the transport regimes using the Binder et al. methodology, as it continues the analysis of trajectories.

After restructuring the paper, the next section that follows is the analysis of the transport regime based on the Binder et al. methodology as suggested by the reviewer.

This section could be made stronger by coupling the delta-theta/delta-T plots with graphs of trajectory clusters (e.g., plots trajectory altitudes as a function of latitude). This would bolster the rather speculative discussion about the meaning of each of the sectors of the Binder plots. This section could get rather long, so it might make sense to give one example (from the springtime flights, perhaps) and place the rest in the supporting materials. This analysis is the part of the current manuscript that is really informative outside of the narrow range of the NETCARE project, as it suggest broader generalities about how transport occurs into the polar dome. I'd like to see it

developed more thoroughly and the conclusions made firmer, perhaps with a concluding paragraph that summarizes the findings from this section.

We included the figures according to the suggestion showing the evolution of the trajectories. As an example, sector 1 of the April 2015 measurements was chosen for those data points inside the polar dome, since it is the most dominant sector. In contrast, sector 4 was chosen for the air masses outside the dome, representing the largest contribution there. For better figure clarity only every 20<sup>th</sup> individual trajectory is plotted. Corresponding figures for the dominating sectors for July 2014 are included in the supplement material.

New figures for trajectory analysis inside the polar dome. Shown is the dominating sector 1 from Fig. 11b in the paper:

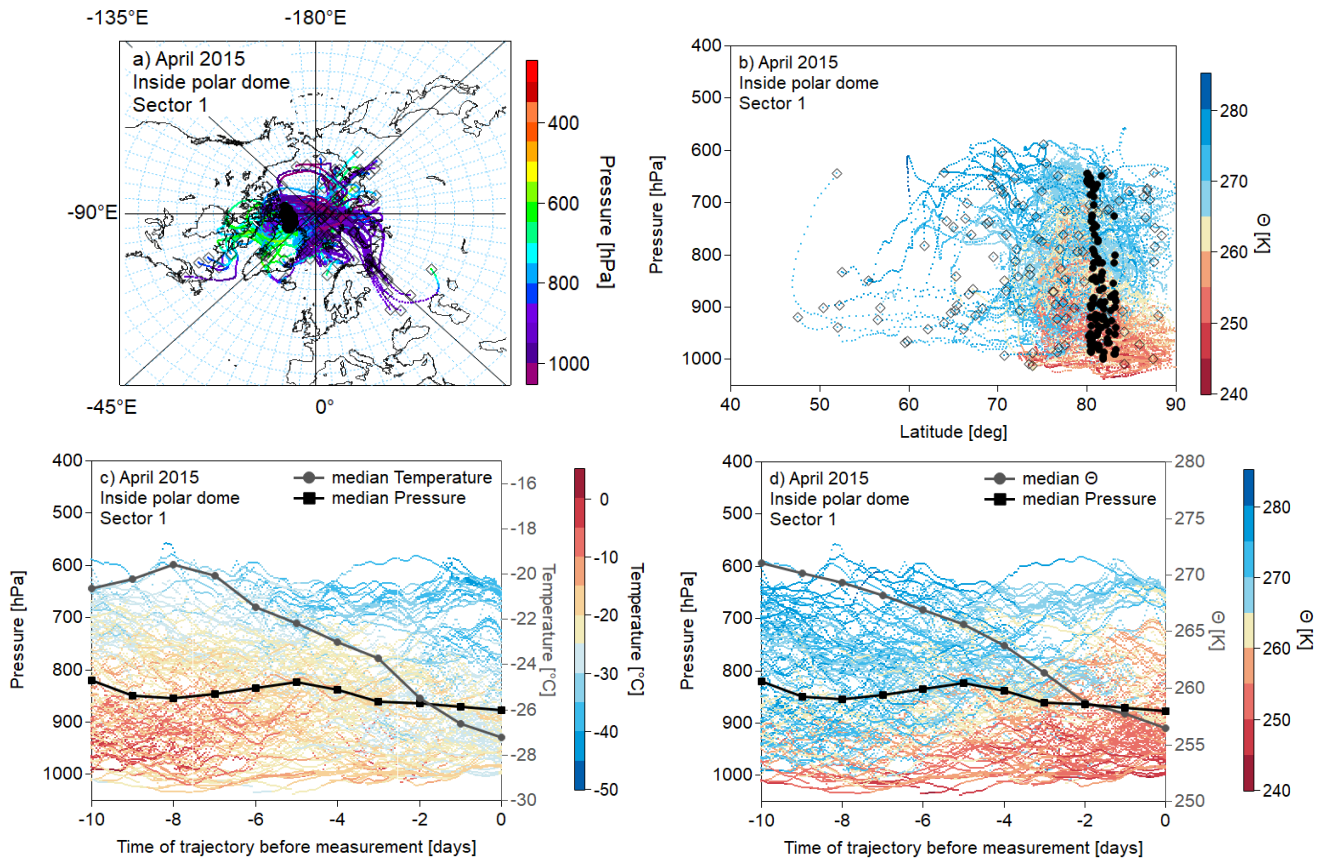


Figure 12: (a) Trajectories of the most dominant sector 1 for air masses inside the polar dome. The color code represents the pressure along the trajectories. (b) The same trajectories as in (a) as a function of pressure and latitude, color coded by potential temperature. In both figures (a) and (b) black circles denote the initialisation point of the trajectory along the flight track. The black open squares show the position of the trajectory 10 days back in time. Figures (c) and (d) show a vertical cross section of the trajectory evolution over the 10 days of travel with the color code denoting the temperature (c) and potential temperature (d). The black line marks the median pressure of the trajectory cluster at the individual time steps and the grey line indicates the median temperature and median potential temperature, respectively. Note that in all figures only every 20<sup>th</sup> trajectory is plotted for figure clarity.

New figures for trajectory analysis outside the polar dome showing the dominating sector 4 from Fig. 11c in the paper:

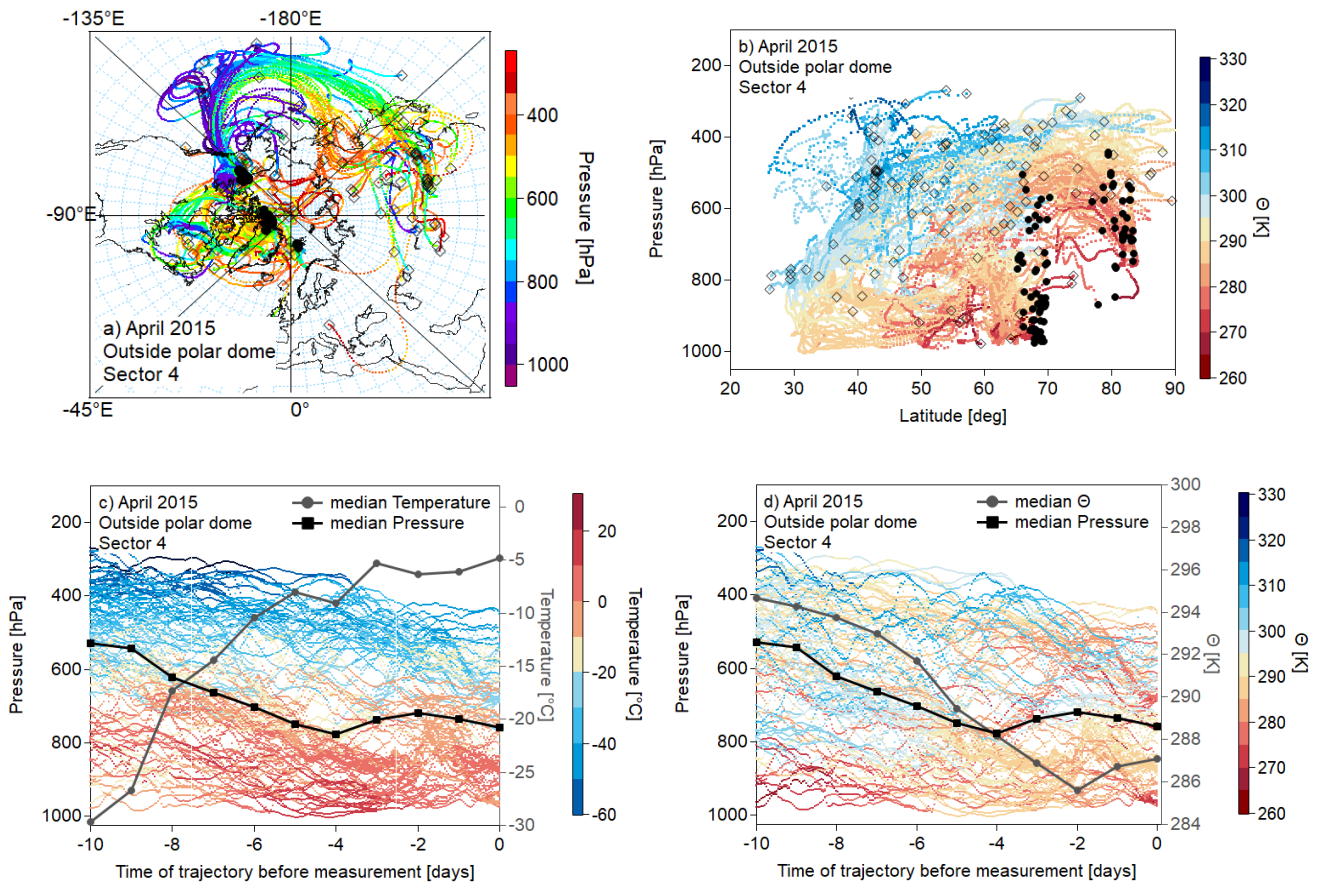


Figure 13: (a) Trajectories of the most dominant sector 4 for air masses outside the polar dome. The color code represents the pressure along the trajectories. (b) The same trajectories as in (a) as a function of pressure and latitude, color coded by potential temperature. In both figures (a) and (b) black circles denote the initialisation point of the trajectory along the flight track. The black open squares show the position of the trajectory 10 days back in time. Figures (c) and (d) show a vertical cross section of the trajectory evolution over the 10 days of travel with the color code denoting the temperature (c) and potential temperature (d). The black line marks the median pressure of the trajectory cluster at the individual time steps and the grey line indicates the median temperature and median potential temperature, respectively. Note that in all figures only every 20th trajectory is plotted for figure clarity.

The following paragraph was added to the manuscript:

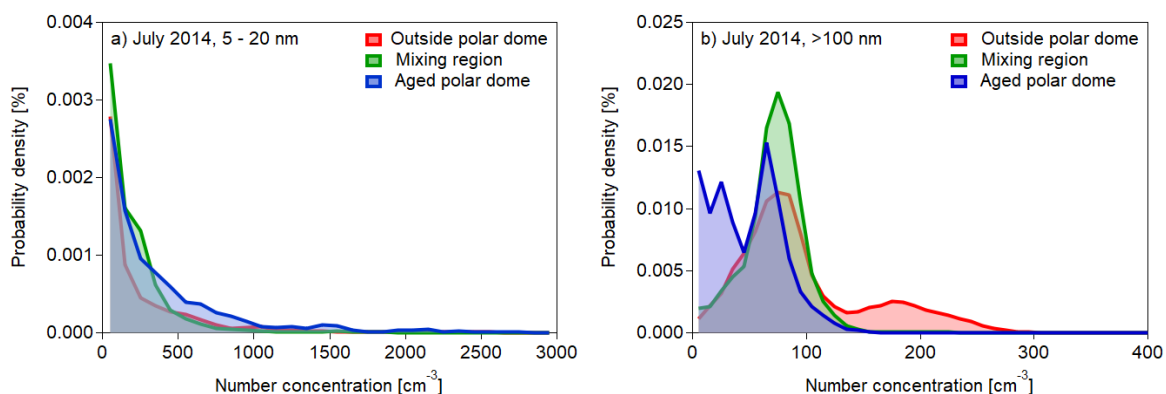
Based on the results from the phase space diagrams we further analyze the trajectories of the individual clusters. This allows for a more detailed analysis of the physical processes along the trajectory. Therefore, we compare the two most dominant sectors for the April 2015 measurements in Figs. 12a-d and 13a-d. Sector 1 is mostly dominated by air masses confined to the central Arctic at all altitude levels (see Figs. 12a and b). The air masses show a weak descent during the 10 days before the measurements but experience a very pronounced decrease in temperature and potential temperature indicated by the evolution of median temperature of the whole trajectory cluster (see Figs. 12c and d). In contrast, the trajectory analysis of the most dominant sector 4 of air masses

outside the polar dome reveals different air streams contributing to the cluster. Air masses originate at different altitudes in the central Arctic, at low level over the Pacific Ocean and from the upper troposphere over Asia. Air masses in this cluster are characterized by a significant increase of median temperature and decrease of median potential temperature indicating a descending trend, which is confirmed by the median pressure decrease over the time of travel. The low-level transport over the Pacific is associated with a low-pressure system over Alaska. Those air masses arrive at the polar dome boundary in the measurement region after experiencing a week net cooling over Alaska.

We conclude that air masses within the aged summertime polar dome are mostly confined to the boundary layer while they experienced a week diabatic warming due to insolation in July 2014 during NETCARE. In the mixing region and outside the polar dome diabatic cooling and a continuous descent is observed. Within the polar dome in April 2015 during NETCARE mostly near-surface processes (diabatic cooling due to the flow over cold surfaces) dominate the recent transport history of air masses in the lower polar dome. Air masses in the upper polar dome experience a very slow descent induced by radiative cooling. Outside the polar dome air masses mostly arrive at higher potential temperatures in the Arctic and experience a continuous slow descent with increasing temperatures but only week diabatic cooling.

Following the transport regime analysis, the next logical section would be to see how these different airmass types are manifested in the pollution loadings. This is currently Sect. 6, which is labelled "discussion". I appreciate the novelty of Fig. 15, but it's very difficult to understand the grey-scale coloring on top of the colored classification scheme. I'd much rather see PDFs of the number concentrations in the 5-20 and 20-100 and >100 size class. Are there also observations of aerosol composition (e.g., BC abundance, composition) that could be added to this section? You've effectively classified the measurements into airmass type; it would be extremely interesting to see how all the available aerosol microphysical and chemical parameters vary within these different airmasses, and compare them with existing literature values.

We thank the reviewer for this point and revised the paragraph in the revised manuscript. We just want to show the relevance of our empirical dome boundary to other applications using aerosol formation as an example. The specific analysis and discussion of aerosol processes is beyond the scope of this paper. We refer to Willis et al. (2019) and Schulz et al. (2019) for a more detailed discussion on aerosol composition within and outside the polar dome. They used the polar dome boundary derived in this study for their analysis. We revised the paragraph on aerosol as follows including a new figure 15:



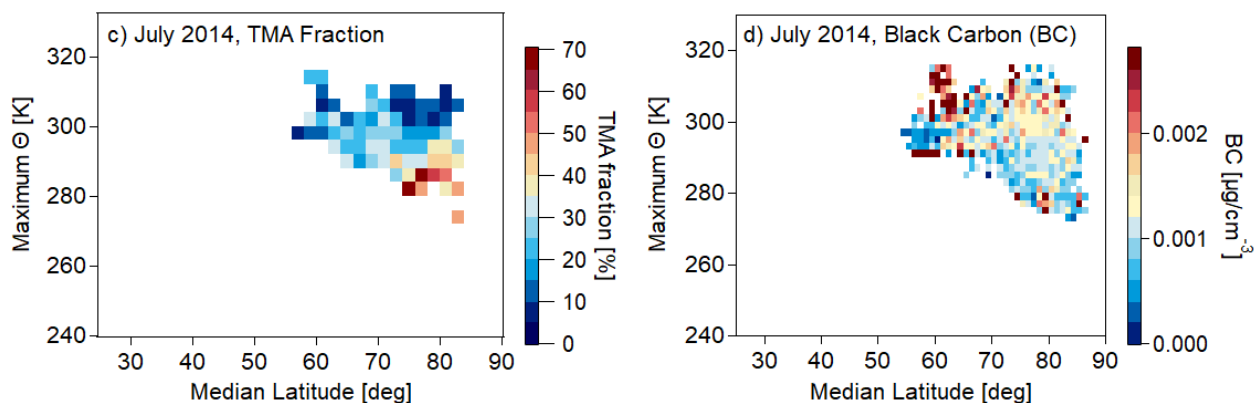


Figure 15: Normalized probability density functions for particles with diameters between 5 and 20 nm (a) and larger than 100 nm for July 2014 (b). The colour code represents the three different regions identified during the polar dome analysis. Panel (c) shows the distribution of the fraction of particles containing trimethylamine (TMA) with respect to the total amount of particles measured by single particle mass spectrometry (Köllner et al., 2017) in the maximum potential temperature and median latitude coordinate system. Note that for the reason of particle spectra statistics the resolution of median latitude and maximum potential temperature interval is reduced compared to the refractory black carbon distribution (BC) shown in panel (d). The enhancement in BC within the polar dome is most probably due to fresh local pollution.

Having defined the polar dome based on trace gas gradients now allows for a more detailed study on aerosol associated with the different air masses. Efficient wet removal and less efficient transport from lower latitudes lead to generally low aerosol concentrations (Stohl, 2006; Engvall et al., 2008), especially within the Arctic lower troposphere during summer. This is consistent with results in Fig. 15a and b. Observations of elevated levels of accumulation mode particles ( $N_{>100}$ ) can be associated with regions outside the polar dome and subsequent transport to the measurement region. In parallel, regions within the polar dome were characterized by  $N_{>100}$  smaller than  $100 \text{ cm}^{-3}$ . In contrary, number concentrations of ultrafine particles ( $N_{5-20}$ ) showed occasionally larger values within the polar dome compared to outside (Fig. 15a), indicating the formation of ultrafine particles occurred within the polar dome region (Burkart et al., 2017). Exemplary for aerosol composition, particulate trimethylamine (measured by single particle mass spectrometry) can be associated with sources within the polar dome (Fig. 15c), consistent with results in Köllner et al. (2017) and Willis et al. (2017). In contrast, the abundance of refractory black carbon can be linked to pollution sources outside the polar dome and subsequent transport to the measurement region (Fig. 15d; Schulz et al., 2019). To conclude, the method introduced in this study is a useful tool to combine Arctic aerosol observations with transport processes and sources within and outside the polar dome region.

Finally, if it's feasible it would be wonderful to extend this analysis to the surface data from the long-term monitoring sites. This would show that the classifications developed here are more broadly applicable. At least an evaluation of whether the approach here is applicable to other cases, or is specific to NETCARE, is needed.

In a follow-up paper to this study we will extend our approach to a variety of different field campaigns in the Arctic covering different seasons and different locations. To demonstrate the applicability of the tracer based diagnostics to a broader data set we analyzed two examples of hourly ground based observations at the Zeppelin Mountain observatory (Ny Alesund, Spitsbergen) (see Fig. R2). Based on CO-CO<sub>2</sub>-scatter plots the signatures and characteristics of both species inside the polar dome during the campaign phases are also found in the respective ground based observations. Note that ground based and airborne observations are in principle affected by different

processes and not necessarily linked. Particularly, the CO and CO<sub>2</sub> data at higher potential temperatures during the airborne campaigns are linked to higher altitudes as evident in both Figs. R1 a and b.

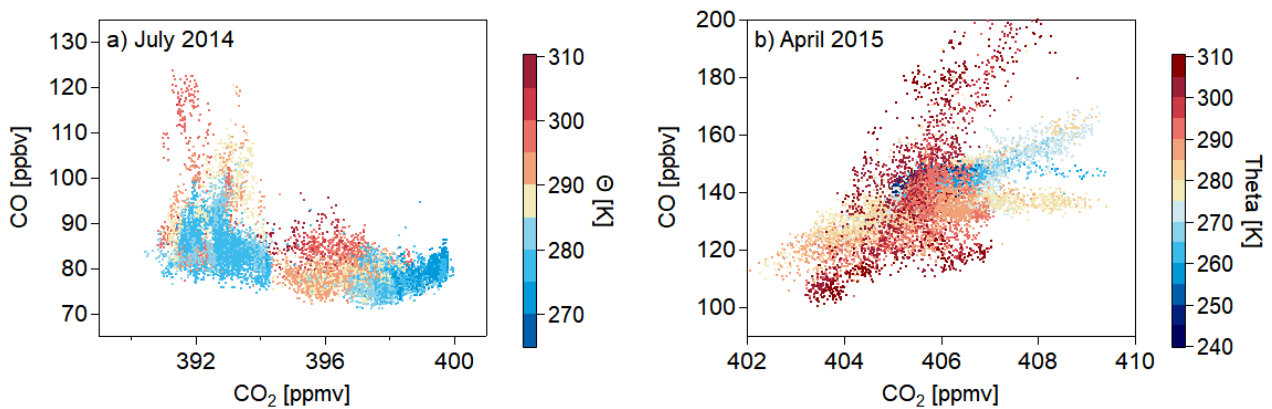


Figure R1 (a): Tracer-tracer scatter plot of all aircraft data points (background + pollution plumes) for July 2014. (b): Tracer-tracer scatter plot of all aircraft data points (background + pollution plumes) for April 2015. The color code denotes potential temperature.

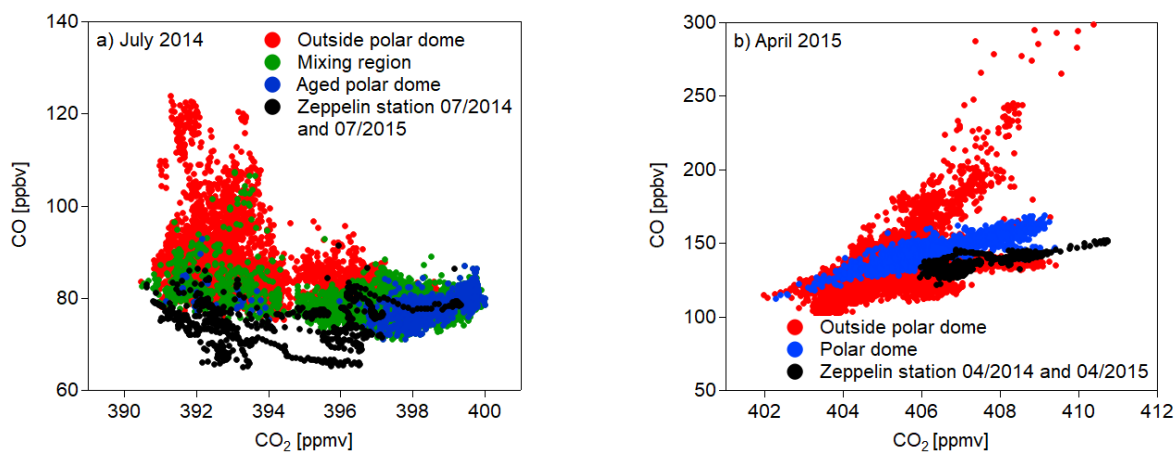


Figure R2 (a): Tracer-tracer scatter plot of all aircraft data points (background + pollution plumes) within the aged polar dome (blue), the mixing region (green) and outside the polar dome (red) for July 2014. (b): Tracer-tracer scatter plot of all aircraft data points (background + pollution plumes) within (blue) and outside (red) the polar dome for April 2015. To separate the different regions the tracer derived polar dome boundaries are used. Boundary values for each region are summarized in Tab. 2 in the paper. The black circles denote the ground based observation data from the Zeppelin mountain observatory (Ny Alesund, Spitzbergen) for the months July (a) and April (b) 2014 and 2015.

Note however, the good agreement at low potential temperatures particularly for the April 2015 case with exactly the same slope and similar absolute values at the station as deduced from the aircraft data as inside dome (see Figs R2 a and b). Based on this analysis we conclude that the Zeppelin observatory was inside the polar dome for April 2014 and 2015 and most probably inside the polar dome with episodes outside the polar dome in July 2014 and 2015. The latter is indicated by the correlation following the characteristics of the mixing region (see Fig. R2a). However, potential temperatures were as low as those observed during the research flights (not shown).

In addition to these larger, structural issues, the manuscript needs some technical correction. The primary author is not a native English speaker, but English-speaking co-authors should step up and give the manuscript a round of thorough copy-editing. Verb tense is not used consistently, which is distracting and sometimes confusing. (Example, p. 17 lines 18-21 go from "now calculated" to "determine" to "finally used".) There are quite a few typos that a spell checker should find, and terms like "surfacenear" instead of "near-surface" are present. This manuscript has a lot of good analysis using an interesting and unique dataset. The CO and CO<sub>2</sub> measurements look spot-on with the long-term ground monitoring network data, which is very encouraging since these airborne measurements can be tricky given the large background. With the suggested restructuring and a tighter focus on how the findings are more broadly applicable, the manuscript should be quite suitable to publish in ACP.

We thank the reviewer for this point. Copy-editing for the manuscript will be done by native English speakers.

Interactive comment on Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2019-70>, 2019.

#### Acknowledgement:

All atmospheric data from Zeppelin are publicly available in the EBAS database (<http://ebas.nilu.no>) and we thank Cathrine Lund Myhre and NILU - Norwegian Institute for Air Research for making the CO and CO<sub>2</sub> observations from Zeppelin available.