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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Analysis of ozone and nitric acid in spring and summer Arctic pollution using aircraft, ground-based, satellite observations and MOZART-4 model: source attribution and partitioning" *by* C. Wespes et al.

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Response to the Anonymous Referee #1:

We thank the referee for his positive general comments about the paper. We acknowledge him for his useful corrections and suggestions, which help us to improve the discussion of our results. Below are our point-by-point responses to his comments, which have been quoted [. . .] before each response.

[Section 2.2, paragraph 2: How did the ARCTAS and ARCPAC O3 and HNO3 measure-

ments compare during the intercomparison flights and do biases between the aircraft measurements influence your conclusions?]

As explained also to the other referee, the preliminary POLARCAT O3 assessment report estimate an O3 deduced relative bias between DC-8 and WP-3D instruments as the sum of: 0.904 (ppbv) + 0.0480*DC-8 O3 (ppbv), with DC-8 O3 taken as an arbitrary reference (http://www-air.larc.nasa.gov/TAbMEP2_polarcat.html). The bias is much lower than the ozone concentrations observed during the flights reported in this paper and do not affect our conclusion. A comparison between DC-8 and WP-3D instruments for HNO3 was also performed and can be found at: http://www-air.larc.nasa.gov/cgi-bin/ic2008r. However, during the comparison period, the ambient levels reported by the DC-8 instrument were within the reported imprecision of the WP-3D instrument making it difficult to assess any bias between these instruments. As suggested by the referee #2, a note on biases between the two data sets has been added in the revised version in Section 2.2 (Model and measurements).

[Section 3, paragraph 2: Figure 4 shows that MOZART-4 significantly overestimates O3 and underestimates HNO3 within the Arctic boundary layer (lowest 0.5km). These biases are briefly mentioned during the discussion of figure 5 but I think that they should be highlighted in the discussion of figure 4 since these biases are systematic. Specifically note that MOZART-4 does not include treatment of the halogen chemistry that is responsible for the extreme ozone depletion events that occur within the Arctic boundary layer. Furthermore, since the Arctic boundary layer tends to be extremely stable and is not resolved very well by numerical weather prediction models it is quite likely that the wet deposition processes that are responsible for HNO3 loss near the surface are poorly simulated (either during transport to the Arctic or within the Arctic boundary layer) leading to the overestimate of HNO3 below 0.5km.]

We thank the referee for pointing out this missing aspect of our analysis. As suggested by both referees, a discussion on the fact that MOZART-4, systematically and simultaneously overestimates O3 and underestimates HNO3 within the Arctic boundary layer

(see Figures 4 and 5 (eg. flights on April 9, 12, and 17) of the manuscript), probably due to the missing halogen chemistry in addition to the misrepresentation of the wet deposition processes in the Arctic boundary layer into MOZART-4, has been specifically added for the analysis of figures 4 and 5 of the manuscript.

[Section 3, paragraph 3: The high correlation between the model and the ARCTAS data most likely reflects the fact that the NASA DC8 sampled the large vertical gradient in the upper troposphere and not necessarily good performance of the model for reproducing variability. The fact that the correlation with the ARCPAC data (which didn't sample the vertical gradient in the upper troposphere due to the lower ceiling of the NOAA P3) is lower supports this conclusion. The fact that the "relative" differences range from -80 to 150% for HNO3 and -40 to 50% for O3 says that the model is not capturing the variability particularly well. The authors need to acknowledge this in the discussion.]

We thank the referee for pointing this out. As suggested by the referee, the interpretation of the high correlation between the model and the ARCTAS observations has been corrected and attributed to the large vertical gradient sampled during the aircraft flight tracks in addition to the fact that the model reproduces the variability gradient of observations in space and time, principally for O3. The values of relative differences range from -80 to 150% for HNO3 and from 40 to 50% for O3, but they are generally close to -10% for O3. Nevertheless, we agree that the model does not fully reproduce the variability of the observations. We found that it generally underestimates the variability range (as seen in Fig.4 of the manuscript) which is principally associated with local or stratospheric influences.

This has been clarified in Section 3 and further discussed in Section 4.1 of the revised paper.

[Section 3, paragraph 5: Please comment on the differences in the compactness of the modeled and observed stratospheric O3/HNO3 relationship. During ARCTAS-A the observations show a relatively broad, but still linear, O3/HNO3 relationship that is

presumably do to different dynamical and photochemical histories of the air masses sampled by the DC8 during the spring when the stratosphere is dynamically active. During ARCTAS-B, the observations show a compact relationship, suggesting a more uniform photochemical history associated with summertime easterly flow and very little stratospheric wave activity. The model shows a very compact relationship for both ARCTAS-A and B, presumably due to the use of climatological values in the stratosphere.]

The suggestions of the referee for the interpretation of the differences in the compactness between the modeled and the observed stratospheric O3/HNO3 relationship in terms of different dynamical and photochemical histories of the air masses sampled along the aircraft flight tracks has been included specifically into the revised paper during the discussion of the Figure 6. As pointed out, the fact that MOZART-4 does not have a complete chemistry in the stratosphere and uses climatologies to constrain numerous species in the stratosphere explains the compact relationship modeled in the stratosphere.

[Section 4, paragraph 1: It seems to me that the signature of transport of anthropogenic HNO3 from Europe and Asia into the Artic is quite high for the ARCPAC measurements. This suggests that the pollution plumes sampled by the P3 were of more recent origin, or didn't experience as significant depositional loss as the more aged European and Asian plumes sampled by the DC8 during ARCTAS. Please comment.]

It is true that Figure 7 of the manuscript shows that the ARCPAC HNO3 contributions from Europe and Asia emissions are larger than for the ARCTAS-A flights. This reflects the different sampling strategies between the two campaigns (as mentioned in the paper), as well as ARCPAC sampling fresher plumes (as suggested by the reviewer). The air masses sampled by the WP-3D aircraft (over Alaska) were more influenced by westerly transport of recent pollution from Asia (anthropogenic and North Asia fires plumes) and from Europe (anthropogenic plumes), while the air masses sampled by the DC8 aircraft (Alaska, North Canada, Greenland) present a stronger influence from

North American anthropogenic pollution. This also highlights the stronger horizontal gradient of anthropogenic HNO3 than anthropogenic O3, as observed in Figures 9 and 10 of the manuscript.

[Section 2.2 paragraph 5: The IASI retrievals should not be averaged over the model grid cell for the comparisons, instead the model should be interpolated in space and time to the IASI retrieval locations. Otherwise, how can the IASI averaging kernels (which are scene specific) be used to accurately convolve the MOZART-4 model profiles?]

For comparisons with MOZART-4 model simulations, the IASI retrievals are averaged over the MOZART grid cells and over the 6-hour window of the model outputs. To take into account the specific scene of each IASI observation, the averaging kernels of the different IASI observations contained in each model grid cell have been used separately to smooth the gridded MOZART profile, and then, for each model grid cell, the average of the smoothed MOZART profiles is calculated. This has been indicated more clearly in the "model and measurements" section (Section 2.2) of the revised version.

[Section 5.1 paragraph 1: Why do the authors consider the column between 300mb and the surface and not something lower? Is 300mb (8km) chosen since this is approximately the mean pressure of the spring-time Arctic tropopause [Hall et al., 2011]? Where does the large daily variance come from? Is this due to variations in the tropopause height?]

The tropospheric column between ground and 300 hPa (typically between the surface and 8km) has been carefully considered both to limit as much as possible the stratospheric and the tropopause height variation influences and to contain the altitude range of maximum sensitivity in the troposphere. The sensitivity of IASI to the ozone profile is characterized by a DOFS ranging from about 2.5 in the polar regions to 3.5 at mid-latitudes and 4 in the tropics, with about one piece of information which is diluted over

the tropospheric range with a maximum peaking around 6-8km altitude for almost all situations (e.g. Figure 2 of the manuscript). As a result, a tropospheric column from ground to 300 hPa is the most relevant for our analysis. This has been clarified in Section 5.1 of the revised paper. The column between ground and 300hPa has also been considered for the analysis of the FTIR partitioning (Section 5.1 of the manuscript) for comparison with the IASI partitioning.

The scatter in the FTIR or IASI data, in Figures 11 and 12 of the manuscript, corresponds to the average of the FTIR or IASI data within the MOZART grid box around Thule or Eureka. As a result, the daily variability (vertical bars in Fig.11 and 12 of the manuscript) represents the variability associated with the scene-specific IASI data within the MOZART grid box. Note also that the number of FTIR observations depends on the day. For some days, only one observation is available and, as a result, no daily variability can be calculated. From Figures 11 and 12 of the manuscript, it can be observed that the large day-to-day variability in both the FTIR and the IASI observations is principally associated with that of the stratospheric contribution. Variability of the anthropogenic sources contributes for a small part of the total day-to-day variability. Similar to the Figure 11 of the manuscript, Figure 1 below represents modeled daily mean of O3 columns from ground to 300hPa at Eureka for 1 April to 13 July 2008 for FTIR observations at Eureka, along with modeled contribution from anthropogenic sources and stratospheric influence, smoothed with the FTIR averaging kernels. The variation in the tropopause height is also represented. The strong day-to-day variability observed in the stratospheric component can be linked to the variation in the tropopause height, explaining a part of the variability observed in the ground-300hPa columns for both IASI and FTIR observations. Similarly, the daily variance (vertical bars in Figure 1 below) could also be partly linked to the daily tropopause height variations. Variation in the thermal vertical structure characterizing the scene-specific FTIR or IASI data could also impact on the daily variance of the observations.

[Section 5.2, paragraph 5: The mean springtime O3 columns below 300mb shown

in Figure 13 are dominated by stratospheric contributions associated with variations in the mean tropopause height and doesn't reflect tropospheric ozone enhancements due to pollution transport into the Arctic. It would be much more informative to show the column below 4km (564mb) so that the ability of IASI to retrieve tropospheric ozone in the Arctic was illustrated. This would also be more consistent with the tagged tracer analysis of the aircraft data since it showed that stratospheric influences dominated above 4km during the Arctic springtime.]

Unfortunately, as discussed above, the sensitivity of IASI is characterized by limited vertical information over the range of the troposphere (about one piece of information in the tropospheric column) with a maximum peaking around 6-8km altitude. This absence of vertical information for tropospheric O3 in the IASI measurements justifies the analysis of a tropospheric column from surface to 300 hPa. Moreover, as discussed in Section 2.2 of the manuscript (see Figure 2 of the manuscript), the vertical sensitivity strongly decreases below 4km down to the surface in the cold Arctic. This is inherent to nadir thermal IR sounding in cases of low surface temperature and low thermal contrast. A column below 4km would not contain information only specific to the lower layers, but it would be affected by information relative to higher layers and by a priori contribution.

[Section 5.2, paragraph 6: Figure 14 is missing so it is difficult to review this discussion.]

It seems that a technical problem occurred during the online publication of the manuscript for the discussion phase. We apologize for the inconvenience. However, Figure 14 is the same as in the pre-print quick review process and is given below).

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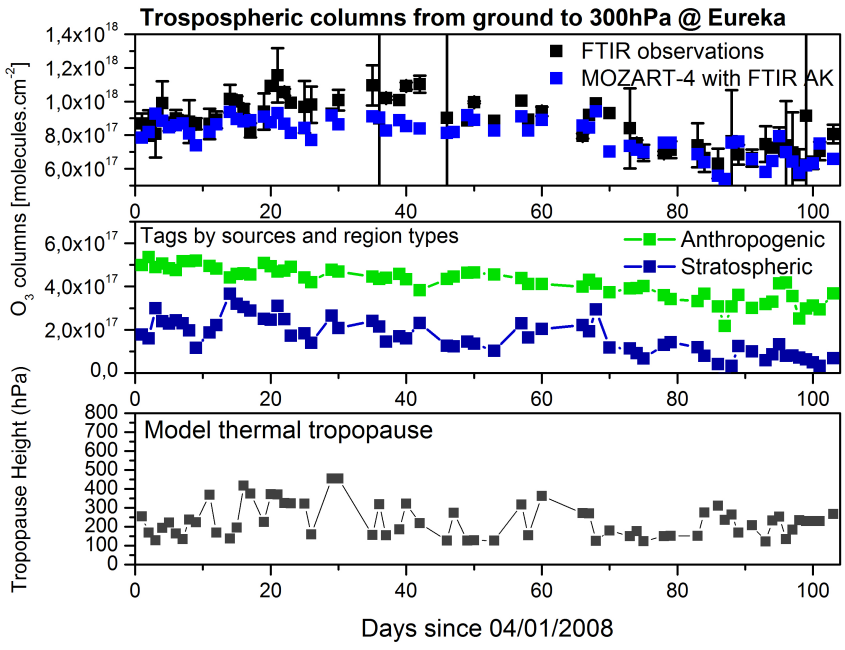

Fig. 1. Top: Daily mean O3 tropospheric columns at Eureka. Middle: Anthropogenic and stratospheric contributions to O3 tropospheric columns. Bottom: Model thermal tropopause height variation (hPa).

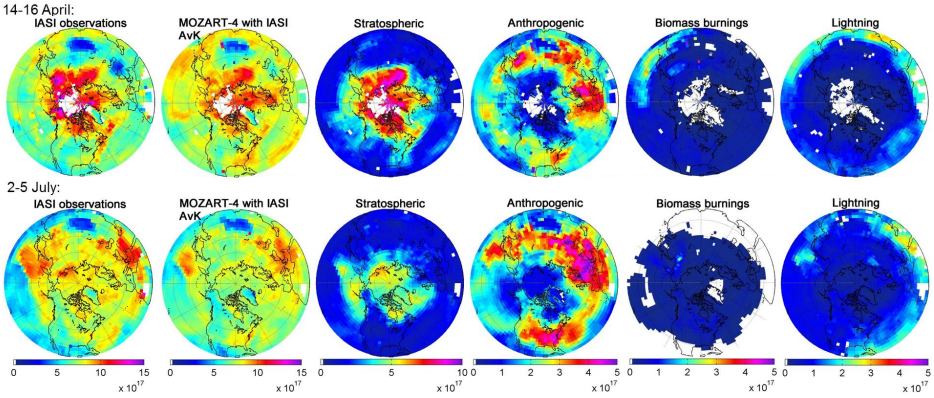

Fig. 2. Mean O3 columns (molecules/cm2) from ground-300hPa over 14-16 April and 2-5 July, observed by the IASI satellite and simulated by MOZART-4 smoothed with the averaging kernels of IASI observations.