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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.

Anonymous Referee #2

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The manuscript entitled "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. provides a detailed comparison between global MOZART-4 model simulations and insitu observations of O3 and HNO3 from aircraft during the 2008 NOAA Aerosol, Radiation, and Cloud Processes affecting Arctic Climate (ARCPAC) and NASA Arctic Research of the Composition of the Troposphere from Aircraft and Satellites (ARCTAS) field missions. These aircraft missions were part of the International Polar Year (IPY) Polar Study using Aircraft, Re-

mote Sensing, Surface Measurements and Models, of Climate, Chemistry, Aerosols and Transport (POLARCAT) program.

The authors show that MOZART-4 is generally able to reproduce the statistics of the in-situ measurements to within 15% and then use MOZART-4 to estimate the relative contributions from anthropogenic, biomass burning, and stratospheric sources of ozone (O3) and nitric acid (HNO3) using tagged tracer techniques. The tagged tracer experiments are then used to relate the MOZART results to the relative contributions as sampled by the NOAA and NASA aircraft. They find that, for the Arctic as a whole, anthropogenic sources dominate Arctic O3 and HNO3 below 400mb with stratospheric sources dominating above 400mb. They further show that the ARCPAC sampling strategy tended to emphasize Asian biomass burning while the ARCTAS sampling strategy tended to emphasize North American anthropogenic sources. Comparisons between MOZART-4 and ground based Fourier Transform InfraRed (FTIR) spectroscopy and satellite Infrared Atmospheric Sounding Interferometer (IASI) O3 retrievals at Thule and Eureka show that the ground based FTIR retrievals are able to measure signatures of Arctic pollution transport better then satellite IASI measurements, which tend to overemphasize stratospheric contributions due to the broader averaging kernels and lower sensitivity to the lower troposphere.

Overall, I find the manuscript to be well written, informative and thorough (with minor exceptions noted below). The MOZART-4 simulation provides a means of integrating the wide range of aircraft, ground based, and remote measurements collected during POLARIS into a coherent picture of Arctic pollution sources during IPY and is a valuable contribution to the POLARIS special issue. Unfortunately, the last figure (figure 14) was not available in the online or downloaded version of the manuscript. This oversight makes it difficult to fully evaluate whether the IASI retrievals are capable of capturing pollution transport into the Arctic, as the authors conclude.

General comments:

ACPD

11, C10568–C10572, 2011

1. Much of the manuscript is devoted to an evaluation of the MOZART-4 model with respect to aircraft measurements, which then provides a foundation for using the model tagged tracer results to interpret the measurements in a broader context. While the evaluation is generally reasonable I think that some specific points need to be emphasized further as outlined below:

Section 2.2, paragraph 2: How did the ARCTAS and ARCPAC O3 and HNO3 measurements compare during the intercomparison flights and do biases between the aircraft measurements influence your conclusions?

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.

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.

ACPD
11, C10568–C10572, 2011

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.

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.

2. The use of tagged tracers, convolved with the instrument averaging kernel, to evaluation the ability of ground based FTIR and IASI retrievals to observe pollution transport into the Arctic is very informative, however, I question the details of their analysis as outlined below:

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?

Section 5.1 paragraph 1: Why do the authors consider the column between 300mb and the surface and not something lower? Is 300mb ($\sim$8km) chosen since this is approximately the mean pressure of the spring-time Arctic tropopause [Hall et al.,

ACPD
11, C10568–C10572, 2011

2011]? Where does the large daily variance come from? Is this due to variations in the tropopause height?

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 ($\sim$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 $\sim$4km during the Arctic springtime.

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

References:

C. M. Hall et al., Tropopause height at 78N 16E: average seasonal variation 2007–2010, Atmos. Chem. Phys., 11, 5485–5490, 2011, doi:10.5194/acp-11-5485-2011

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 23707, 2011.