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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 #2:

First of all, we thank the referee for his useful comments on the study, which have helped us clarifying some points and improving the manuscript. Below are our point-by-point responses to his comments and suggestions. Before each response, the reviewer comments have quoted between [. . .]. Corresponding information and corrections have been added to the revised version of the manuscript. Technical comments are also

included in the revised version.

Specific Comments:

Attribution of model biases relative to aircraft:

[Section 3, Paragraph 3: You note that the model has a bias relative to the aircraft measurements. Later, you use the model to separate the contribution of various source regions in your tagged analysis to the pollutant concentrations at the aircraft locations, and some of those contributions are of the same order as your model bias (eg. 5-15% for ozone). How does the model bias relative to observations affect the interpretation of the tagged tracer results?]

We thank the referee for pointing out this missing aspect of our analysis. The quantitative partitioning for the smallest contributors (<15% of the total ozone, see Fig.8 of the manuscript) to the total ozone concentrations should be looked at with more caution. Some words of caution have been added in the revised version of the manuscript (Section 4.1), in the discussion related to the figure 7, to insist on the qualitative character of the smaller contributions.

However, for the ARCTAS measurements, the underestimation of O3 and HNO3 concentrations is principally attributed to a mix of underrepresentation of local pollution sources (North American anthropogenic pollution, Californian and Canadian fires) and stratospheric influence (see comment below), depending on the altitude and the campaign period. During ARCPAC, the underestimation is principally attributed to an underrepresentation of Russian fire emissions, Asian and European anthropogenic pollution. These contributions represent together the largest contribution to the total O3 or HNO3 (much larger than 15% for O3, see fig.8 of the manuscript). The model bias relative to observations should not significantly affect our conclusion related to the analysis of the relative partitioning along the aircraft tracks and over the Arctic. However, we agree that the bias of the model relative to the aircraft measurement should be pointed out when discussing the partitioning of the tagged tracer. It has been added in Section 4.1

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of the revised version of the manuscript.

Note that the model bias relative to observations has been taken into account when comparing the model results with the FTIR and the IASI observations.

[Section 4.1, Paragraph 4: What motivates the equating of variability with the cause of bias? Just because a particular source type is more variable does not necessarily mean it is the most likely cause of the bias. For example, imagine your model is missing a factory near your observation location that emits NO_x at a constant rate. This would not necessarily contribute much to the variability in the model, but it does contribute to a bias. Conversely, some sources will naturally be more variable than others, but that does not mean they are biased. Also, in this paragraph you mention the Russian fires from 2008. There is some literature concerning model representation of these fire events during ARCTAS (Fisher et al., 2010, ACP, "Source attribution and interannual variability of Arctic pollution in spring constrained by aircraft (ARCTAS, ARCPAC) and satellite (AIRS) observations of carbon monoxide" and references therein).]

We agree with the referee that the bias between the model and the aircraft observations, which has been principally attributed to an underestimation of anthropogenic, fires and stratospheric influences, cannot directly be inferred from an analysis of the associated variability. Nevertheless, when separating data by anthropogenic, stratospheric or fires influence, we observe that the model specifically underestimates concentrations from local pollution (anthropogenic and fire pollution) in the lower/middle altitudes, while it underestimates concentrations largely influenced by stratosphere in the higher altitudes. For example, during ARCTAS-B, when considering data strongly influenced by North American anthropogenic pollution and Californian fires (note that these two contributions are generally coincident in the ARCTAS observations), we found the median of the differences between observed and simulated O3 to be around -6.3%. Similarly, if we consider data strongly influenced by Canadian fires during ARCTAS-B, we calculate a median difference of -9.9%. Considering together data influenced by these local emissions (North American anthropogenic pollution, Californian and Cana-

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dian fires) which represent the dominant contributors to O3, we observe a negative median difference of -7.5%. When separating data strongly influenced by stratospheric fluxes during the ARCTAS-A spring phase, we calculate a negative median of -28%. The negative values calculated for specific influenced data largely explain the value reported when considering all the ARCTAS-B observations (Fig.4 (a) of the manuscript). These specific concentrations influenced by local emissions or stratospheric influence are also characterized by the largest variability. Even if, as indicated in the paper and following the high correlation coefficients (R) between model and observations for the ARCTAS data sets, we are confident that the model captures the variability gradient of observations in space and time, we found that it also generally underestimates the variability range (horizontal bars in Fig.4 of the manuscript) which is principally associated with local or stratospheric influences. This could, for example, partly result from the coarse horizontal and vertical resolution of the model which cannot capture the larger variability associated with the local emissions and the stratospheric fluxes and also from the interpolation of the MOZART-4 results (from 6-hour averages) to the flight path (from 1–min averages). These model uncertainties could also explain part of the bias between the observations and the simulations. Underestimation of both concentrations and variability into the model can hardly be dissociated even if they do not necessarily result from the same model errors.

The equating of the variability with the attribution of the bias to underestimations of specific tagged contributions has been corrected and better explained in the revised text.

Following the suggestion of the referee, Fisher et al. (2010) has been referred in the discussion related to the model representation of the Russian fires. Note, however, that Fisher et al. (2010) used a different fire inventory than the one we used here.

[Section 3, Paragraph 4: You note two reasons that stratospheric air masses are underrepresented in the model. Plume dilution will be a factor, but how true is it that the constraint of long-lived species in the stratosphere to a climatology may be causing a

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bias in the stratospheric plumes in the model? Is this because 2008 was atypical for stratosphere-troposphere exchange? It may be worth including a mention of how the MOZART stratosphere is parametrized in this study in particular (is SYNOZ used, or is the downward ozone flux with the NCEP-GFS meteorology doing a good enough job on its own?).]

As described in details in Emmons et al. (2010), MOZART-4 does not have a complete chemistry in the stratosphere. It uses a climatology for specifying several species in the stratosphere, including O3, CO, CH4, HNO3, NOx, which is not specific for 2008. The mixing ratios are set to zonal mean climatologies above 50hPa coming from MOZART-3 simulations (MOZART-3 being an extension of MOZART-2 into the stratosphere and mesosphere (Kinnison et al., 2007)) and then, they are constrained by relaxation toward the climatology down to the tropopause. The stratospheric flux of ozone is not constrained by the SYNOZ representation. When the MOZART simulations are driven by the NCEP/GFS analyses, it is not necessary to use SYNOZ to simulate ozone concentrations in the upper troposphere (Emmons et al., 2010). Nevertheless, due to the constraints to the stratospheric mixing ratios mentioned above and to the reanalysis meteorological datasets (here the NCEP/GFS analyses), MOZART-4 is not well adapted for studies in the tropopause region/stratosphere (Emmons et al., 2010; Grewe et al., 2002). Some discussion of the missing stratospheric chemistry and parameterizations has been added in the Section 2.1 (MOZART-4 model description).

[Section 3, Paragraph 4: From Figure 5, it seems as though when the aircraft encounters low ozone values near the surface (eg. flights on April 9, 12, and 17), there is a simultaneous peak in the nitric acid observations. This contributes to your model underestimate of nitric acid. How significant is the missing bromine chemistry to nitric acid through the changes in the oxidative state of the Arctic boundary layer? Can you speculate as to the direction (or magnitude) of the bias this might introduce in your resulting simulated nitric acid concentrations?]

Sensitivity simulations are required to evaluate the impact of the missing bromine

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chemistry to the modeled nitric acid. Since the bromine chemistry is not represented into the MOZART-4 model, we cannot evaluate the magnitude of its contribution to the production of nitric acid, by comparing model results with and without the bromine chemistry. Nevertheless, as suggested by the referee #1, the systematic and the simultaneous overestimation of O3 and underestimation of HNO3 in MOZART-4 (see Figures 4 and 5, e.g. flights on April 9, 12, and 17 of the manuscript,) could result from both the missing halogen speciation and the misrepresentation of the Arctic boundary layer into MOZART-4, including the missing bromine-induced extreme ozone depletion and an overrepresentation of the wet deposition processes leading to an underestimation of HNO3 near the surface. A note on the systematic and simultaneous overestimation of O3 and underestimation of HNO3 into MOZART-4 has been added during the discussion of figures 4 and 5 of the manuscript.

IASI description and interpretation:

[Section 1, Paragraph 3: The description of the IASI instrument is better suited to the "Model and measurements" section.]

Following the suggestion of the referee, the description of the IASI instrument has been moved to the Section 2.2.

[Section 2.2, Paragraph 4: Where does the FORLI-O3 system obtain its a priori information? This is pertinent to your analysis in that where IASI has low sensitivity, so the retrieval will be relying on its a priori information (eg. Figure 12 shows the a priori contribution in April being second-most significant after the stratosphere). Also, in the IASI instrument description, it may be worth discussing the satellite's overpass times and effects of the diurnal cycle in the IASI observations. If the overpass is at roughly the same time of day, how does the change in the thermal structure in the Arctic atmosphere between spring and summer affect the retrieval? Do the IASI observations capture the same diurnal or seasonal cycle as the FTIR observations?]

As described in Scannell et al. (2011), the a priori information (a priori profile

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and a priori covariance matrix) used for the retrieval of O3 is built from the Logan/Labow/McPeters climatology (McPeters et al., 2007) which is a combination of data from the Stratospheric Aerosol and Gas Experiment II (SAGE II; 1988–2001), the Microwave Limb Sounder (MLS; 1991–1999) and data from balloon sondes (1988–2002) to represent the best approximation of the atmospheric state. Note that a single O3 a priori profile and variance-covariance matrix are used. This has been added in the "model and measurements" section of the revised manuscript. Some words describing the satellite's overpass have also been added in the IASI instrument description section. However, as it has been indicated in the paper, we only consider in our analysis the daytime observations of the O3 IASI measurements. The daytime FTIR nadir observations are characterized by a higher surface temperature and a higher thermal contrast, with, as a consequence, a better vertical sensitivity to the troposphere (Clerbaux et al., 2009; Boynard et al., 2009), which is important for studying source pollution and partitioning. The variations of surface temperature and thermal contrast do have an impact on the averaging kernels associated with concentration retrievals, with a better sensitivity to the tropospheric column during the summer than during the spring, as shown by the a priori contribution in Figure 12. This influence of the surface temperature and thermal contrast impacts on the monthly variations observed from the IASI nadir, with, for example, a larger contribution of the anthropogenic pollution observed by IASI during the summer while this contribution, as observed by FTIR instrument, is larger during the spring (see Figure 11 and 12). On the contrary to the IASI nadir satellite observations, the ground-based FTIR observations are not dependent on the surface temperature and thermal contrast, and the variations of the stratospheric contribution can only partly mask the variations of the tropospheric ozone. The influence of surface temperature and thermal contrast on the analysis of tropospheric O3 IASI observations, and the reason for only considering the daytime observations in our analysis have been indicated more clearly in the "Model and measurements" Section 2.2. We also would like to point out that the FTIR observations only provide daytime solar absorption measurements, when the sun is above the horizon (for the Eureka and

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Thule sites, such measurements can be made approximately from February to October (e.g. Lindenmaier et al., 2010)). As a consequence, diurnal variations cannot be detected from the FTIR observations.

[Section 5.2, Paragraph 2: The interpretation of the various influences here to the IASI column is not as unambiguous as the text implies. The averaging kernels for IASI in Figure 2 extend well above the tropopause, and it is not clear whether the stratospheric influence you see here is actually stratospheric air transported into the troposphere below 300 hPa or if the averaging kernels are smearing down information from higher altitudes into the surface-300 hPa column. Also, there is a large contribution from the a priori, probably from where the averaging kernels are small in the lower troposphere. This happens to be where the non-stratospheric sources are most important (fires and anthropogenic). Can the mismatch between IASI and the model really all be attributed to the model's underestimate of the stratospheric source? How well does the FORLI-O3 a priori capture transport from fires and anthropogenic pollution near the Arctic surface?]

As mentioned in the manuscript during the discussion of Figure 12, the observations from IASI at Thule and Eureka are dominated by the stratospheric contribution, resulting from a higher sensitivity of IASI in the higher tropospheric layers where the stratospheric influence represents the largest contributions (Fig. 7 and 10 of the manuscript). Importantly, there is about one piece of decorrelated information over the range of the troposphere with a maximum sensitivity peaking at around 6-8 km for almost all atmospheric situations (e.g. Figure 2 of the manuscript). However, over the coldest polar region, the IASI sensitivity is lower, implying some dilution of information from higher altitudes into the ground-300hPa columns. Note also that the spring-time Arctic tropopause height is close to 300 hPa (see Figure 1 in the responses to referee #1) explaining also some contribution from lower stratosphere. The stratospheric influence coming from stratospheric air transported into the ground-300hPA columns or resulting from a troposphere which is not fully decorrelated from stratospheric information can-

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not be discriminated at this stage. The IASI measurements are also affected by the a priori information because of a lower vertical sensitivity of IASI in the lowest layers. As a consequence, IASI detects smaller contributions from anthropogenic pollution and fires than the FTIR instruments. As discussed, from Figure 12 of the manuscript, a sharp decrease of the a priori contribution between April and July observations is observed. The larger contribution during the spring reflects a lack of sensitivity probably due to both smaller thermal signal above ice-covered surfaces and smaller thermal contrast in the cold boundary layers. The smaller a priori contribution during the summer reflects a better sensitivity with, as a consequence, a larger contribution from the non-stratospheric sources. The impact of the IASI sensitivity and a priori profile are taken into account when smoothing of the model profiles to the lower vertical resolution of the observations for the purpose of comparison. As a result, they cannot explain the mismatch between IASI and the model. As discussed in Section 5.2 of the manuscript, the high differences (averaged negative bias of around 20%) between observations and model datasets over the Arctic are not all attributed to model's underestimate of the stratospheric source. They are partly attributed to the underestimation of the stratospheric contribution in the model, to the dilution on the model spatial scale ($\sim$140 km) compared to IASI spatial scale ($\sim$12 km at nadir), to the tendency of IASI retrievals to overestimate the tropospheric ozone columns by $\sim$10% in the Northern latitudes (Boynard et al., 2009).

Discussion of regional influences and transport:

[Section 4.1, Paragraph 5: A few different factors contribute to the relative increase of Asian influence with altitude. Isentropic transport along the so called polar dome (eg. Stohl, 2006, JGR, "Characteristics of atmospheric transport into the Arctic troposphere") makes the Arctic upper troposphere more accessible than the lower troposphere to pollutants emitted at high potential temperature (such as those that originate in east/southeast Asia). The partitioning of reactive nitrogen at higher altitudes in the Arctic tends to favour the reservoir species (HNO3, PAN) over NOx. An interesting

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question is whether the relative importance you see in Asian HNO3 is due to an overall greater abundance of Asian reactive nitrogen, or whether the partitioning from that source favours HNO3 over other reservoir species.]

As discussed in Section 4.2 (Figures 9 and 10), both Asian and North American emissions dominate concentrations over the American sector of the Arctic between the surface and 400 hPa, through lifting to the middle troposphere and transport towards higher latitudes, associated with WCBs. It has been identified as the major transport pathway for pollution originating from North America and Asia (Stohl, 2006). This might explain the relative increase of Asian influence with altitude along ARCTAS-B flight tracks. Similarly to Figure 8 of the manuscript, Figure 1 below presents the partitioning of principal sources of PAN (left) and NOy (right) along the ARCTAS summer flight tracks. We observe simultaneously a relative increase of Asian HNO3 and reactive nitrogen (NOy = NOx+PAN+HNO3) abundances with altitude. The relative increase of Asian influence with altitude is also observed for PAN. However, contrary to the partitioning of HNO3, the relative contributions of PAN and NOy from Asia are not significantly larger than the relative contributions from other anthropogenic individual sources (American and European) at higher altitudes, suggesting that the partitioning from that source favors HNO3 over other reservoir species with altitude.

[Section 4.2, Paragraph 1: Your discussion of Asian HNO3 seems inconsistent with the previous paragraph. Here it is short-lived due to washout and uptake on dust and contributes little to the Arctic columns, whereas in Section 4.1 it was transported far from the NOx source and was therefore long-lived, was a candidate for pollution transport, and was significant in the Arctic upper troposphere. Can you reconcile these two descriptions?]

In the manuscript, we indicate that, generally and relative to anthropogenic O3 which shows horizontally smoothed distributions, anthropogenic HNO3 presents a strong horizontal gradient overall the troposphere, probably due to washout or to the uptake of HNO3 on dust or in comparison with O3, making it shorter-lived and therefore, less

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available than O3 for transport into the Arctic. In the previous section (Figure 8 of the manuscript), we specifically discuss about the increasing relative contribution of Asian anthropogenic HNO3 in higher altitude along the ARCTAS summer flight tracks, making HNO3 a candidate for transport of pollution in higher altitudes, through transport with WCBs, in comparison with lower altitudes. It was not discussed in comparison with O3 which is also transported into higher altitudes with WCBs. It has been rewritten in the revised version to avoid misunderstanding.

[Section 4.2, Paragraph 1: Your discussion of North American ozone also conflicts with what is described in the previous section, and what is shown in Figures 7 and 8. The North American source dominates at lower altitudes, so how can its contribution be qualified as "small"? Does this reflect a bias in the sampling of the ARCTAS flights, since most of them were in the North American sector?]

During discussion of Figures 7 and 8 of the manuscript, we point to the fact that the local pollution sources, principally North American anthropogenic pollution, dominate the contribution to tropospheric O3 in the lower altitudes along the aircraft fight tracks, while, in section 4.2, we discuss the partitioning over the Arctic (north of 60°N) in the ground-300hPa columns where the pollution from North America is found to contribute only a small amount to the Arctic pollution (around 1.5$\times$1017 molecules/cm2 on average for O3). As discussed in the last paragraph of Section 4.2., this difference reflects the fact that the partitioning based on the aircraft track flights is generally different from that for the entire Arctic (See table 2 of the manuscript). The aircraft sampling patterns influence the partitioning analysis, with, for example, an overrepresentation of the North American anthropogenic contribution ($\sim$10% for O3) in the lower/middle troposphere, in comparison with partitioning from the entire Arctic.

[Throughout: The literature is not always consistent when it refers to "the Arctic" as a region. I don't think it is mentioned explicitly anywhere in this paper either, but be clear what you mean by it, whether it is north of 60N, north of the Arctic circle (66N), or north of 70N, or something else.]

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In this paper, we consider the Arctic region as the region north of 60N. In the Section 4.2: Partitioning over the Arctic, the indicated values are related to the values observed north of 60N. The definition of the Arctic region, such as it is used in this paper has been added. Moreover, the use of terms 'Arctic' or 'northern latitudes' has been corrected throughout the manuscript in a consistent way.

[Section 2.1, Paragraph 2: The description of your model emissions is misleading. They cannot be "constant in time" because Table 1 has different totals in spring and summer. Do the emissions change each month and have constant values during each month?]

As described in Section 2.1, the anthropogenic emissions have constant values with no monthly variation. Table 1 represents total value (Tg) of surface global or regional NOx emitted during the ARCTAS spring (1-19 April) and summer (18 June - 13 July 2008) campaign periods. The ARCTAS spring period corresponds to a period of 19 days while the summer period corresponds to a period of 26 days, explaining why the value given for the summer period is larger than this given for the spring period. The total NOx emissions for the two periods are represented in Tg (not in Tg/day).

[Section 2.2, Paragraph 2: Were the ozone and nitric acid instruments on the two aircraft platforms intercompared? Make a note of possible biases between the two data sets.]

As describe in Brock et al. (2011), instruments comparisons were performed during separate coordinated flights with the NASA DC-8 and the NOAA WP-3D at the same altitude. Results from the comparison can be found at: http://www-air.larc.nasa.gov/TAbMEP2_polarcat.html for O3 and at: http://www-air.larc.nasa.gov/cgi-bin/ic2008r for HNO3. An O3 deduced relative bias between DC-8 and WP-3D instruments is estimated as the sum of: 0.904 (ppbv) +0.0480*DC-8 O3 (ppbv), with DC-8 O3 taken as an arbitrary reference. The bias is much lower than the ozone concentrations observed during the flights reported in this paper and do not af-

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fect our analysis. For HNO3, the ambient levels reported by the DC-8 instrument during the comparison period were within the reported imprecision of the WP-3D instrument making it difficult to assess any bias between the instruments.

As suggested by the referee, a note on biases between the two data sets has been added in the revised version in Section 2.2 (Model and measurements).

[Section 2.2, Paragraph 3: Could you clarify whether the DOFS value quoted here is the mean for the campaign? Is the mean DOFS significantly different between the spring and summer campaigns? Between the two sites? What is the range of the FTIR DOFS?]

Generally, as indicated in Section 2.2, the IASI DOFS varies over the globe from about 2.5 in the polar regions to 3.5 at mid-latitudes and up to 4.5 in the tropics, with about one full piece of information in the troposphere peaking around 6 km for almost each atmospheric situation encountered. Some differences between the spring and the summer periods can be observed in the mid- and polar latitudes over the land, reflecting higher sensitivity during the summer (e.g. DOFS of about 3-4 during the summer vs. 2.5-3.5 during the spring in the N.H.), induced by higher surface temperature and higher thermal contrast. The DOFS are marginally different between the Eureka (80N, 86W) and the Thule (77N, 69W) sites, which are located at similar latitudes. The FTIR DOFS are also similar between the two sites and range from about 5.8 to 6.5 over the spring and the summer campaigns. This has been clarified in the revised version of the paper.

[Section 5.2, Paragraph 5: What is the evidence that the larger errors are due to water vapour lines? Is there a paper you could reference here?]

Surface temperature and water vapor profile are retrieved concurrently with ozone profile, inducing “cross-state” errors between retrieved multiple parameters. The “cross-state” errors result from the effect of simultaneous retrieved states on a particular state (e.g. cross-state errors on ozone profile retrieval due to the simultaneous retrieval of

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water vapor profile) and contribute, along with the measurement and the smoothing errors to the total retrieval error of ozone. The total retrieval errors on the retrieved tropospheric columns are strongly variable on the globe. Higher errors found in the equatorial belt can be explained by the impact of water vapor lines which strongly interfere with O3 features. Larger total retrieval errors found above the equatorial belt and attributed to strong overlapping water vapor lines have already been published in Wespes et al. (2009), which describes the retrieval of nitric acid from IASI. A paper dedicated to the retrieval of IASI spectra with the FORLI fast radiative transfer model/inversion software and to a complete description of the FORLI products in term of retrieval performances at a global scale is in progress (Hurtmans et al., submitted).

[Table 1: Is lightning included under "Other sources"? It should be separated into its own row, since you are showing explicit contributions from lightning in later figures.]

As indicated in Table 1, which presents global and regional NOx surface emission sources, other sources only include emissions from soils and from aircraft. As suggested by the referee, emissions of NO from lightning have been added separately into its own row in the revised version of the paper.

[Figure 8: Can you comment on how well the NOy partitioning agrees with the aircraft observations from ARCTAS? Alvarado et al., 2010 (ACP, "Nitrogen oxides and PAN in plumes from boreal fires during ARCTAS-B and their impact on ozone: an integrated analysis of aircraft and satellite observations") had difficulty reproducing this feature in the Arctic (too much HNO3, not enough PAN). How might a bias in the partitioning affect your analysis?]

Figure 2 below presents the vertical distribution of O3, NOx, HNO3, PAN, averaged over 0.5 km layers, of the ARCTAS-B aircraft observations on 29 June – 10 July, along with the MOZART-4 model values interpolated to the flight tracks (Horizontal bars represent the standard deviation associated with the observations - the correlation coefficient and median of the differences are also indicated.). The differences relative to

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the observations, expressed as a percentage, are shown on the right panels for each campaign. Note that for comparisons between model simulations and aircraft observations, the MOZART-4 model results are sampled along the flight track making use of 1-min average aircraft measurements, instead of 15-min average aircraft measurements used in Alvarado et al. (2010).

The better matches between the ARCTAS-B DC-8 observations and our MOZART-4 results at lower altitudes might suggest that the FINN fire emissions, version 1 (Wiedinmyer et al., 2010) may be more accurate than the FLAMBE emissions for the local Canadian emissions sampled at lower altitude and that the anthropogenic emissions from the 2006 inventory of Zhang et al. (2009) may be more appropriate than those used in GEOS-Chem (van Donkelaar et al., 2008) to represent Asian and North American anthropogenic pollution. Note also that, on the contrary to Alvarado et al. (2010), we do not observe simultaneous large overestimation of HNO3 with underestimation of PAN over all the altitudes of the observations. We also observe very small differences between GEOS-Chem model results and MOZART-4 model results for O3, suggesting that the different anthropogenic and fires emissions, chemistry schemes used for the both models have a limited impact on ozone simulations. Further model intercomparison exercises are needed to evaluate the impact of different parameters used in the models on NOy partitioning and to evaluate the impact of the rapid formation of PAN on ozone concentrations in fresh plumes (Alvarado et al., 2010). However, we agree that a bias in the NOy partitioning might affect the simulated HNO3 and O3 concentrations. It has been added in the revised version in Section 4.1 when discussing the different model errors possibly explaining the bias between observations and simulations.

Technical Comments:

[A number of small corrections to grammar and spelling are listed below, along with suggested additions and clarifications of the text.]

According to the suggestions of the referee, the technical corrections have been in-

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cluded in the revised version of the manuscript. Below are our responses to the specific comments about suggestions and clarifications into the text.

[Table 2: This table would be more readable if the entire Arctic and aircraft sampling values were separated into different columns instead of using the slashes.]

It has been done in the revised version.

[Page 23729, Lines 5-7: Is there good reason for the precision of these contributions to change? I appreciate that there are more IASI observations than there are FTIR, but isn't the limit on your precision how well your model can do the source segregation? Compare here with Page 23727, Lines 16-18 or all of Page 23725.]

The precision of the contributions to total O3 has been corrected in Section 5.1.

[Figure 5: For Flight 10, the altitude scale begins at -5km, where no good aircraft should go.]

We plot the altitude scale from -5km to make the altitude of the observations not superimpose the O3 or HNO3 concentrations for clarity.

[Figure 8: Make all the horizontal scales the same (0-100%) to allow for easier visual comparisons.]

It has been done in the revised version of the manuscript.

[Page 23724, Line 11: "According to the ARCPAC observations..." Do you mean the simulations along the ARCPAC flight tracks?]

Indeed, we mean simulations along the ARCPAC flight tracks and not the aircraft observations. This has been corrected in the revised version.

[Figure 9: Make a note in the caption that the colour scale of the nitric acid plots changes.]

It has been specifically indicated in the caption of Figure 9 in the revised version of the

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manuscript.

[Throughout: The word “tags” should be replaced with “tagged tracers” for consistency and clarity.]

This has been changed in the revised version.

[Figure 14: Figure 14 has disappeared!]

Apparently, a technical problem occurred during the online publication of the manuscript for the discussion phase. We apologize for the inconvenience. Figure 14 is the same as in the pre-print quick review process.

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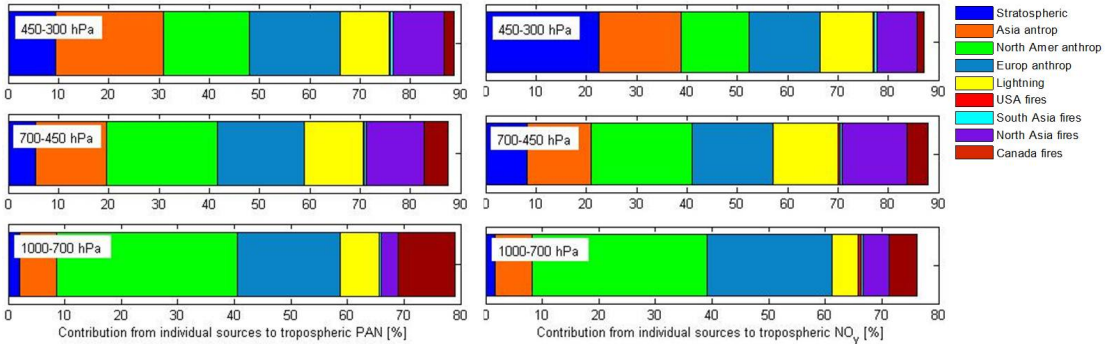

Fig. 1. Partitioning of principal sources of PAN (left) and NOy (right, approximated by NOx + PAN + HNO3) in three altitude bands along the ARCTAS summer flight, simulated by MOZART-4, in % of the total.

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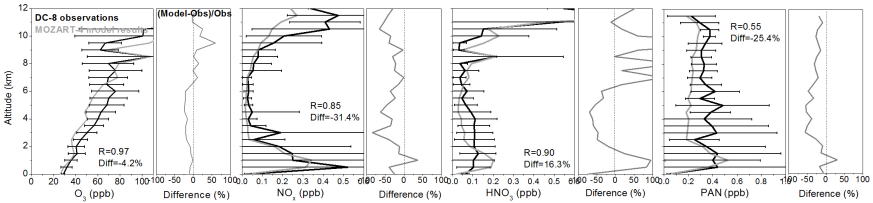

Fig. 2. Vertical distribution of O3, NOx, HNO3 and PAN observations during ARCTAS-B on 29 June – 10 July, averaged over 0.5 km altitude and compared to the MOZART-4 model values (from 6-hour averages).

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