

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 #1

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General Comments

This manuscript presents a model-based analysis of ozone and nitric acid in the Arctic in the context of the ARCTAS and ARCPAC aircraft campaigns. Data from the aircraft are used to evaluate the model, which is then used to provide information about sources of pollution to the Arctic. The manuscript also presents the first use of ozone retrievals from the IASI satellite instrument over the Arctic.

The methods used in this work are commonly accepted, and the study builds on pre-C9709

vious works that focus on Arctic pollution, presenting data from the aircraft campaigns and the IR platforms in a comprehensive and clear manner. However, I have a number of specific comments that I would like the authors to address to improve the manuscript before publication.

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?
- **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, ARC-PAC) and satellite (AIRS) observations of carbon monoxide” and references therein).

- **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 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?).
- **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?

• IASI description and interpretation

- **Section 1, Paragraph 3:** The description of the IASI instrument is better suited to the “Model and measurements” section.
- **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

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(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?

- **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?

• 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

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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 NO_x. An interesting 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.

- **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 NO_x 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?
- **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?
- **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 60°N, north of the Arctic circle (66°N), or north of 70°N, or something else.
- **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?

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- **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.
- **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?
- **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?
- **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.
- **Figure 8:** Can you comment on how well the NO_y 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?

Technical Comments

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

- **Page 23709, Line 15:** Change “stratospheric-contaminated” to “stratosphere-contaminated”.

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- **Page 23709, Line 25:** Switch the order of “ground-based FTIR” and “IASI satellite sounder” to make it clear that the ground-based measurements are at these two sites. IASI measurements have a broader range.
- **Page 23710, Line 13:** The phrase “during spring and summer 2008” is repeated twice.
- **Page 23711, Line 9:** Change “a period” to “periods.”
- **Page 23712, Line 5:** Change “despite their” to “despite its.”
- **Page 23712, Line 8:** The ARCPAC acronym has already been defined on Page 23710, Line 12.
- **Page 23712, Line 13:** Change “on October 2006” to “in October 2006.”
- **Page 23715, Line 19:** Change “WP-3” to “WP-3D.”
- **Page 23716, Line 12:** The subject and predicate in this sentence do not match: “They reveals a DOFS...”
- **Page 23717, Line 4:** Suggest adding: “... comes from the a priori *at these altitudes*.”
- **Page 23718, Line 7:** Change “previously” to “previous.”
- **Page 23719, Line 4:** Change “course” to “coarse.”
- **Page 23719, Line 6:** Change “performances” to “performance.”
- **Page 23722, Line 26:** Change “transpacific” to “trans-Pacific.”
- **Page 23723, Line 4:** Change “fires emissions” to “fire emissions.”

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- **Page 23723, Line 6:** Suspect that “latitudes” here should be changed to “altitudes.”
- **Page 23723, Line 12:** Change “non-complete” to “incomplete.”
- **Page 23724, Line 2:** Suggest adding: “Fig. 9 shows *simulated* spring and...”
- **Page 23724, Line 4:** There is an extra bracket in there. Also on lines 8 and 10.
- **Page 23724, Line 11:** “According to the ARCPAC observations...” Do you mean the simulations along the ARCPAC flight tracks?
- **Page 23724, Line 28:** These distributions are shown in Fig. 10a and 10b, not Fig. 9a and 9b.
- **Page 23725, Line 25:** Change “gradient” to “gradients.”
- **Page 23725, Line 27:** The Figure numbers here are reversed (Fig. 8 shows the aircraft flights, and Fig. 10 shows the entire Arctic).
- **Page 23726, Line 2:** Change “is observed” to “are observed.”
- **Page 23726, Line 6:** Change “from entire Arctic” to “from the entire Arctic.”
- **Page 23726, Line 24:** The averaging kernels are shown in Figure 2, not Figure 1.
- **Page 23726, Line 25:** Be consistent about the use of negative signs when designating a negative bias (compare here to Page 23728, Line 15).
- **Page 23727, Line 8:** Equation could be set on a separate line.
- **Page 23727, Line 24:** Change “associated to” to “associated with.”

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- **Page 23728, Line 8:** No comma here.
- **Page 23728, Line 12:** The averaging kernels are shown in Figure 2, not Figure 1.
- **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.
- **Page 23729, Line 11:** Change "amplitudes up of" to either "amplitudes up to" or "amplitudes upwards of." It is not clear which is meant.
- **Page 23730, Line 23:** Add "lightning" to list of contributions shown. This is assuming Figure 14 (which is missing here) is still the same as in the pre-print version.
- **Page 23730, Line 25:** Change "concentrations" to "columns."
- **References section:** Some of the article titles containing chemical names are missing subscripts (for example, O3 instead of O₃)
- **Table 2:** Change "principal sources to" to "principal sources of" in caption.
- **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.
- **Figure 3:** (a) and (b) are used twice ambiguously in this caption. Perhaps use (a) through (d) instead?
- **Figure 4:** The σ symbols in the caption have disappeared.

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- **Figure 5:** For Flight 10, the altitude scale begins at -5km, where no good aircraft should go.
- **Figure 8:** Make all the horizontal scales the same (0-100%) to allow for easier visual comparisons.
- **Figure 9:** Make a note in the caption that the colour scale of the nitric acid plots changes.
- **Figure 13:** The multiplier on the colour scale ($\times 10^{15}$) has disappeared.
- **Figure 13:** Add a superscript in the caption to the units molecules cm⁻².
- **Figure 14:** Figure 14 has disappeared!
- **Throughout:** The reference "Jacob et al., 2009" should be "Jacob et al., 2010" as it is in the References section.
- **Throughout:** The word "tags" should be replaced with "tagged tracers" for consistency and clarity.

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

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