

## Response to Reviewer1

### General comments

This manuscript is well written. However, the authors sometimes use imprecise formulations or make statements of too general nature. Comments are given in the specific comments section.

**Reply: We carefully addressed all the general and specific comments.**

Some more detailed information on the model characteristics (e.g. how aerosol aging is handled, what types of emission inventories were used etc., see specific comments) are needed to make the study reproducible.

**Reply: We added a detailed model description including emissions of CO and BC, oxidant fields for CO chemistry, and BC aging process and its dry/wet depositions. See reply to specific comments below.**

The study is partly based on the tagged CO tracers. However, the overall validation of GEOS-5 CO results with DC-8 CO measurement data is only conducted for the total CO and not the tagged species. There is data such as e.g., acetonitrile from the DC- 8 measurements to test the overall BB contributions. Also, for anthropogenic CO the SO<sub>2</sub> and SO<sub>4</sub> data could be used. Some more data comparison or at least discussion on the quality of the tagged CO species is needed before interpreting flight data with model runs.

**Reply: Good point. GEOS-5 uses 5 CO tags to track pollution origins to the Western Arctic. Based on the reviewer's suggestion, we analyzed the correlation of modeled biomass burning CO with observed CH<sub>3</sub>CN. We also analyzed the correlation of modeled fossil fuel CO with observed CH<sub>2</sub>Cl<sub>2</sub> since CH<sub>2</sub>Cl<sub>2</sub> is a well-known fossil fuel tracer. See new Figures 4-6 and the discussion in section 3.2 of validation of the model capability in characterizing source attribution.**

It is very useful that the authors address a number of tracer species and not only CO. However, most of the discussion is focused on the evaluation of the CO model/measurement inter-comparison while such analysis for SO<sub>2</sub>, BC, and particulate sulfate is rather neglected.

**Reply: In this paper, the model is used to investigate regional Western Arctic pollution. Therefore, it is critical to demonstrate that the model can reproduce measurements statistically over the studied spatial and temporal domain. With this in mind, we added Figure 3 to compare aerosol BC vertical distributions between GEOS-5 simulation and DC-8 measurement, similar to what we did for CO in Figure 2. For a reader's reference, we also gave a more detailed model-observation comparison of BC along each flight track (Figures S2 and S4) and a composite of measured BC concentration compared to model results (Figure S6). The correlation coefficients between model and measurement are given in the figures.**

**We removed the content of SO<sub>2</sub> and SO<sub>4</sub> for a cleaner and more concise discussion.**

In the end, it is not clear what the authors conclude from the model performance except that the emission inventories might be responsible for the discrepancies between model results and measurements. A statement on whether further model development or improvement especially regarding non CO species is needed to better represent pollution sources to the Western Arctic is missing.

**Reply: We augmented the analysis of the model performance for non-CO species (i.e. BC) studies in this work. Based on the reviewer's suggestion, we added discussions regarding the processes that need to be improved for BC simulations. Please see**

**section 2.2 (chemistry and dry/wet deposition), 2.3 (emission), 3.1 (paragraphs 4-5 for model BC evaluation), and the last paragraph of the conclusion.**

Specific Comments

p. 8825, l. 20: "Midlatitude pollutants are often . . . a region also vulnerable to climate change. . ." This statement is too general. It is not clear which pollutants the authors refer to and how climate change is linked to these pollutants. There is a large number of publications that are suitable to underline the specific sensibility of the Arctic to climate change that can be referred to in addition to Chapin et al.

**Reply: The sentence has been re-written as "Mid-latitude pollutants, including very short-lived species such as aerosols, are often transported to the Arctic (Barrie, 1986; Weber et al., 2003)."**

p. 8825, l. 24: Tell which campaigns were conducted (ARCTAS A, B, ARCPAC, POLARCAT-France etc.) with respective citations.

**Reply: We added the campaigns (e.g. ARCTAS, ARCPAC, POLARCAT-France, POLARCAT-GRACE, YAK-AEROSIB) and their corresponding references.**

p. 8826, l. 11: What do you mean by contradictory?

**Reply: We deleted "contradictory identification of" to avoid confusion.**

p. 8826, ll. 10-14: This statement sounds like industrial emission regions were shifted around in the mid-latitudes. A more precise formulation stating that the regions of significant emissions changed. Why only within the past 30 years? The clean air acts in the US and Europe were partly enacted prior to the 1980s which led to a significant change in the composition of emissions due to e.g., low sulfur fuels. Also, not only the most significant emitter regions changed, but also the fuel type or mix changed over the last decades. Also, include a reference for the statement.

**Reply: We changed the statement to "Emissions from Europe have declined and anthropogenic activities in Asia have greatly increased in the past two decades (e.g., Streets et al., 2009). Recent investigations suggest that Asian pollution sources have become significant to Arctic pollution; their degree of importance is still being debated (Fisher et al., 2010; Koch and Hansen, 2005; Shindell et al., 2008; Singh et al., 2010; Stohl, 2006). The uncertainty of the origin of Arctic pollution is partly due to complicated pathways for transport of air from mid-latitudes to the Arctic (Fuelberg et al., 2010) and partly due to the uncertainties in estimating emission amounts and types in different regions."**

p. 8826, l. 23: rather Western Arctic than the Arctic as a whole

**Reply: Done.**

p. 8827, l. 7-9: what do you mean by "are not sufficient for the Arctic regional wide pollution during ARCTAS-B"? Not sufficient for what? Explaining the observations, yielding a comprehensive study for larger geographic areas. . .

**Reply: Simply averaging aircraft measurements is not sufficient to represent the pollution level region wide in July. We deleted this sentence to avoid any confusion.**

p. 8827, l. 10: some more explanation of the two categories "long-range transport and boreal forest fires" is needed. What kind of long-range transported pollution do you mean? Does this include both, emissions from anthropogenic and biomass burning sources? By especially stating "boreal forest fires" do you mean "local" boreal forest fires?

**Reply: The sentence was changed to “This study focuses on assessing the impact of long-range transport of mid-latitude anthropogenic pollution and boreal forest fires, both local and foreign, on Western Arctic atmospheric composition during both ARCTAS-A and ARCTAS-B.”**

p. 8827, l. 17: “sulfate containing aerosols (SO<sub>4</sub>)”, it is probably not externally mixed

**Reply: Like most bulk mass CTM models, SO<sub>4</sub>, as well as BC, OC, dust, and sea salt, is mixed externally in our model. We only took into account BC in this study.**

p. 8827, l. 18: “was conducted” instead of “provides insight into the objectives of this paper.”

**Reply: Done.**

p. 8827, l. 19: “. . . is not subject to dry and wet removal processes but can be oxidized by reaction with the hydroxyl radical (OH). . .”

**Reply: Done.**

p. 8828, ll. 3-4: delete “this pair of tracers is also useful in pursuing our objectives.”

**Reply: Done.**

p. 8828, ll. 8-10: What about SO<sub>2</sub> processing in clouds? Conversion to particulate sulfate is very rapid and results in a small SO<sub>2</sub>:SO<sub>4</sub> ratio and the aerosols are not necessarily washed out if no precipitation event occurs. In such cases the air mass can be very young and have a smaller ratio than an aged air mass which only underwent dry chemistry.

**Reply: Deleted the content regarding SO<sub>2</sub> and SO<sub>4</sub>.**

p. 8828, l. 22: in section 2.1 you are also referring to SO<sub>2</sub> but it is not mentioned in the section title, also specify what you mean by aerosols because it can mean a number of characteristics like number density, mass concentration, chemical speciation etc. “2.1 CO, SO<sub>2</sub> and chemical aerosols species from aircraft measurements”

**Reply: Changed the title to “CO and BC concentrations from aircraft measurements”.**

p. 8829, ll. 4-14: The information about the in situ measurements is inconsistent. State temporal resolution, accuracy or estimated error for all instead of writing “high . . . temporal resolution” or not stating it at all.

**Reply: Added information of time resolution, accuracy, and references for the measurements of SP2 BC, CH<sub>2</sub>Cl<sub>2</sub>, and CH<sub>3</sub>CN.**

p. 8829, l. 14: DeCarlo et al., 2006 would be the original HR-ToF-AMS paper, or Cubison et al. 2011 who operated the instrument

**Reply: No long relevant.**

p. 8830, l. 21: What does primarily mean? If you used other sources they should be referred to as well. If not then “primarily” can be deleted.

**Reply: Deleted “primarily”. The source discussion has been moved to section 2.3.**

p. 8831, l. 21: in Figure 1 you only use longitudinal coordinates referring to the East. In the text and Fig. 2 captions you use Western coordinates.

**Reply: The Western coordinates in Fig. 8 (previously Fig. 2) and the text have been changed to be consistent with labels in Figure 1.**

p. 8832, l. 1: What do you mean by “Ideally, we would like..”? Aircraft measurements hardly represent a wide geographic area or seasonal or multi-year variance with their data. They only provide a snapshot in time and space?

**Reply: We mean that, if possible, we would like to analyze Western Arctic pollution using measurement data directly. But such a study sometimes is not straightforward since aircraft measurements have spatial and temporal limitations. The model is useful for a study of large scale pollution.**

p. 8832, ll. 7-10: This paragraph seems to be out of place. It would fit better in section 2.3.

**Reply: This sentence has been deleted for concision.**

p. 8833, l. 1 2: Insert the waypoints after “the US-Canada border” and “the Arctic near Greenland” to make it easier for the reader to find on the graph what you mean.

**Reply: We added “(waypoints 1-3)” after “the US-Canada border” and “(waypoints 8-10)” after “the Arctic near Greenland”.**

p. 8833, l. 8: What are surrounding areas of Asia? The surrounding areas would cover large parts of the Northern Hemisphere, specify.

**Reply: We explained the surrounding areas specifically in the next sentence in the text: “Specifically, the southern plume originated from Asia and Eurasia and the northern plume from northeast Asia and Labrador Sea (Fuelberg et al., 2010, <http://fuelberg.met.fsu.edu/research/arctas/traj/traj/html>).**”

p. 8833, l. 20: instead of stating “right above” the altitude in hPa would inform the reader better about the location of the plume.

**Reply: We added “the Asian anthropogenic peaks at 300-400 hPa, higher than the 400-500 hPa peak for the boreal biomass burning” in the paragraph.**

p. 8834, l.2: “the tracer concentrations. . .” instead of “aerosol concentrations”, because you refer to SO<sub>2</sub> afterwards as well.

**Reply: The paragraph has been removed since we no longer discuss SO<sub>2</sub> tracer.**

p. 8834, l. 4: What does greatly mean? Can you give a delta value?

**Reply: The paragraph has been removed, see reply above.**

p. 8834, ll. 5-13: This paragraph is repetitive. Suggestion: “Checking the model CO mixing ratios . . . showed that the southern plume. . .” and drop the speculation in lines 5-9 where you essentially say the same. Was there any precipitation along the northern pathway? Also, what do you want to state with “, which is comparable to BC’s lifetime.”?

**Reply: The paragraph has been removed, see reply above.**

p. 8834, ll. 16-20: Not clear what you are trying to say. First you state that the air mass was clean within that period, then you list the source regions without discussing why there was no pollution.

**Reply: The previous figure 4 and the corresponding discussion including this paragraph in the manuscript on ACPD have been deleted based on the comments of the second reviewer.**

**(In the manuscript on ACPD, we want to say the atmosphere before waypoints 5 is “relatively” clean compared with the atmosphere after waypoints 5 (see figure 4). However, the atmosphere before waypoints 5 still contains pollution from the regions,**

**but in a very low concentration.)**

p. 8835, ll. 6-8: It is unclear why these ratios were not calculated, discussed and shown in Fig. 3 for the first flight example as well. This is inconsistent. I suggest providing these data as well for the first flight.

**Reply: The paragraph has been removed, see reply above.**

p. 8835, l. 14: SO<sub>2</sub> emissions from biomass burning are significantly lower than from fossil fuel emissions. “emitted from fossil fuel and from biomass burning to much lesser extent. . .”

**Reply: The paragraph has been removed, see reply above.**

p. 8836, l. 12: “by the GEOS-5 model along the flight tracks.”

**Reply: Done (see section 3.1 last paragraph).**

p. 8835, l. 18 – p. 8836, l. 6: You need to elaborate on how GEOS-5 handles the aging of BC aerosol and what specifically might be the problem. Is BC aerosol aging too slowly? Also, are the Shindell model comparison and your runs comparable? Did you use the same emission inventories? Can the overestimation of BC mass concentration be due to other reasons than BC removal processes? Some more discussion is needed.

**Reply: This paragraph has been deleted (see reply to p. 8834, ll. 16-20), but we added the discussion of the BC emissions (section 2.3), aging (section 2.2), and dry/wet depositions (section 2.2) in GEOS-5 for the ARCTAS study and compared the GEOS-5 performance with the results from GEOS-Chem in these sections.**

p. 8837, l. 1f: What do you mean with “vice versa”? It is very unlikely that the measurements have missed high CO mixing ratios along the flight track while a model simulation might actually miss a plume.

**Reply: We use “vice versa” simply indicating that sometimes model CO is high while measurement is low. This could happen when the model does something wrong. For example, if the model places a fire plume by mistake in grid box A, next to its real location in grid box B along the flight track, the model CO will be high and the measurement will be low at box A, while the opposite will be true at box B.**

p. 8837, ll. 9-13: “The comparisons indicated that the model agrees well with the measurements along the flight track when the compared samples are averaged within 1 km vertical bins. Explain why this type of study is particularly important. What do you mean by “give us some confidences”? What type of GEOS-5 studies are you referring to? What confidences do you mean?”

**Reply: This type of evaluation demonstrates that the model captures the measurements when the tracers are averaged first within 1 km vertically. Our purpose is to use the model to investigate region wide pollution, without chasing individual plumes. Therefore, this comparison, combined with the other evaluations conducted in the paper, gives us some confidence in using GEOS-5 in the study of region wide Arctic pollution.**

p. 8838, l. 6: Elaborate what you refer to with transport efficiency, the term is not clearly defined.

**Reply: The transport efficiency is defined to be a statistic fraction of a tracer mass in plumes compared to its mass in a subset of plumes that basically do not experience wet removal along transport [please refer to Matsui et al. [2011a] for more details]. As a result, it can be regarded as a measure of the impact of wet removal process during**

**transport. To avoid confusion, we remove the phrase “transport efficiency”.**

p. 8840, l. 14: “. . .model simulations of CO.” AIRS wasn’t used for the other tracers.

**Reply: No long relevant.**

p. 8840, l. 19: “It will be misleading. . .” replace by: “Aircraft data alone cannot provide a comprehensive and representative picture of Arctic pollution in July due to the limitations in spatial and temporal coverage.” The use of aircraft data as such is not misleading, this statement is too strong and would mean that the data is useless. You probably meant to say that aircraft data alone cannot provide information that is representative enough.

**Reply: Thanks for pointing this out. Please refer to the new description for the application of the measurement and model in the second paragraph of Section 5.**

**“The model reveals that the ARCTAS DC-8 measurements are representative of regional Arctic pollution in the spring (April, ARCTAS-A) due to relatively homogeneous tracer distribution. The aircraft data alone, however, are insufficient to provide a comprehensive and representative picture of Arctic pollution in the summer (July, ARCTAS-B) because the flights targeted local fire plumes.”**

p. 8841, l. 21f: for the SO<sub>2</sub>/SO<sub>4</sub> ratios the units are not correct

**Reply: No long relevant.**

Technical Comments

p. 8825, l. 2: We present “the” analysis of. . .

**Done.**

p. 8825, l. 7: explain the abbreviation “AIRS”

**No long relevant.**

p. 8826, ll. 12: “. . .transport of air from mid-latitudes to the Arctic. . .”

**Done.**

p. 8827, l. 14: the abbreviation AIRS hasn’t been fully explained yet in the text

**No long relevant.**

p. 8827, l. 27: “sources”

**Done.**

p. 8828, l. 4: “has” instead of “have”

**Done.**

p. 8828, l. 17: “using” instead of “paying careful attention to”

**Done.**

p. 8828, l. 24: close parenthesis after link

**Done.**

p. 8831, l. 1: “produced by the NASA. . .”

**Done.**

p. 8831, ll. 9-11: “three anthropogenic emission source regions from..., and two biomass burning emission source regions from. . .”

**Done.**

p. 8831, l. 17: “identify”

**No long relevant.**

p. 8831, ll. 20 21: “panels”

**Done.**

p. 8832, l. 18: “are shown in Fig. 3a. . .”

**Done.**

p. 8833, l. 9, 21: “northern”, and sometimes you use a capital letter for “Northern” sometimes

you don't

**Changed all to "northern".**

p. 8833, l. 12: "in the right column"

**Done.**

p. 8833, l. 14: "biomass burning pollution contributions, "

**Done.**

p. 8834, l. 2: "But contrary to the aerosol enhancement. . ."

**Done.**

p. 8834, l. 5: "This observation. . ." instead of "interesting phenomenon"

**No long relevant.**

p. 8834, l. 15: "different air masses which were clean before. . ." p. 8834, l. 22: "with contribution from . . ." p. 8835, l. 15: "with only few percent of direct particulate sulfate emissions." p. 8835, l. 20: "over the US" p. 8835, l. 17: "The April campaign (ARCTAS-A) focused on the . . ."

**No long relevant.**

p. 8836, l. 27ff: "The areas covered by the July campaign (ARCTAS-B) experienced strong influence from local fires so that the observed CO mixing ratios partly exceeded 1500 ppbv.

**Done.**

p. 8838, l. 2: ". . . confined to the lower and middle troposphere."

**Done.**

p. 8838, l. 6ff: ". . . less efficient summer transport with 4

**Done.**

p. 8838, l. 20ff: what do you means with "The model simulated CO results sampled same as flight measurements. . ."?, and ". . . is shown as red line and the model result . . . is shown as green shaded. . ."

**The sentence has been rephrased.**

**"The probability distribution function (PDF) of modeled total CO was calculated for each of the five source groups, four of which are shown in Fig. 9. The EUFF tag was excluded from the figure since the sampled total CO was almost never dominated by the tagged European pollution. The PDFs from observation and model along flight tracks are also shown in the figure as an additional model evaluation."**

p. 8838, l. 28ff: " both ASFF and BOBB in July which is in good agreement with the results from Fig.2a, b. "

**Done.**

p. 8839, l. 2: what do you mean by "could", occasionally, has happened in a specific incident?...

**No long relevant.**

p. 8838, l. 7: "in April" instead of "from"

**Done.**

p. 8839, l. 21: "Arctic" instead of "North Pole"

**Done.**

p. 8839, l. 23: again, what do you mean with "could"?

**The sentence has been rephrased.**

p. 8840, l. 5 "during ARCTAS-B"

**Done.**

p. 8840, l. 8: "can" instead of "could"

**Done.**

p. 8840, l. 24: ". . .(accounting for. . .)"

**Done.**

Fig. 2: In the text and Fig. 1 captions you use NBBB and BOBB, in the Fig. 2 legends it's turned around, the red line Obs CO is not dotted in c and d but in a and b.

**Done.**

Make sure Figs. 3 and 4 are large enough in the print version.

**Sure.**

Fig. 3 a, b the waypoint numbers are too small and sometimes can't be distinguished from other black lines on the plot (same for Fig. 4). Captions: (c) is the curtain "plot". NABB trace in Fig. 3 d right is hardly visible.

**No long relevant.**

Fig. 3 and 4 give all panels in the figures a letter for clearer reference.

**No long relevant.**