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Interactive comment on "Investigation of source attributions of pollution to the Western Arctic during the NASA ARCTAS field campaign" by H. Bian et al.

Anonymous Referee #1

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Refeere's comments on Title: Investigation of source attributions of pollution to the Western Arctic during the NASA ARCTAS field campaign Author(s): H. Bian et al. MS No.: acp-2012-109 MS Type: Research Article

This work makes a contribution to the understanding of what type of emission source regions and source types are responsible for the spring and summer pollution in the Western Arctic. The authors use DC-8 in situ data to validate the GEOS-5 model results for a variety of gas phase and particulate tracers. In addition the authors use tagged CO species to provide detailed information on pollution sources. This work also makes the point that aircraft measurements cannot always provide representative

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measurements for a larger area due to the spatial and temporal limitations.

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.

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.

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 SO2 and SO4 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.

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 SO2, BC, and particulate sulfate is rather neglected.

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.

Specific Comments

- p. 8825, I. 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.
- p. 8825, I. 24: Tell which campaigns were conducted (ARCTAS A, B, ARCPAC, POLARCAT-France etc.) with respective citations.
- p. 8826, l. 11: What do you mean by contradictory?
- p. 8826, II. 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.
- p. 8826, l. 23: rather Western Arctic than the Arctic as a whole
- 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...
- 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?
- p. 8827, l. 17: "sulfate containing aerosols (SO4)", it is probably not externally mixed
- p. 8827, l. 18: "was conducted" instead of "provides insight into the objectives of this C2948

paper."

- p. 8827, l. 19: "... is not subject to dry and wet removal processes but can be oxidized by reaction with the hydroxyl radical (OH)..."
- p. 8828, Il. 3-4: delete "this pair of tracers is also useful in pursuing our objectives."
- p. 8828, II. 8-10: What about SO2 processing in clouds? Conversion to particulate sulfate is very rapid and results in a small SO2:SO4 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.
- p. 8828, I. 22: in section 2.1 you are also referring to SO2 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, SO2 and chemical aerosols species from aircraft measurements"
- p. 8829, II. 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.
- 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
- 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.
- 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.
- 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?

- p. 8832, II. 7-10: This paragraph seems to be out of place. It would fit better in section 2.3.
- 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.
- p. 8833, I. 8: What are surrounding areas of Asia? The surrounding areas would cover large parts of the Northern Hemisphere, specify.
- p. 8834, l. 20: instead of stating "right above" the altitude in hPa would inform the reader better about the location of the plume.
- p. 8834, I.2: "the tracer concentrations..." instead of "aerosol concentrations", because you refer to SO2 afterwards as well.
- p. 8834, l. 4: What does greatly mean? Can you give a delta value?
- p. 8834, II. 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."?
- p. 8834, II. 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.
- p. 8835, II. 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.
- p. 8835, l. 14: SO2 emissions from biomass burning are significantly lower than from fossil fuel emissions. "emitted from fossil fuel and from biomass burning to much lesser extent..."
- p. 8836, l. 12: "by the GEOS-5 model along the flight tracks."

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- 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.
- 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.
- p. 8837, II. 9-13: "The comparisons indicated that the model agrees well with the measurements along the flight track when the compared samples are averaged within 1km 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?
- p. 8838, l. 6: Elaborate what you refer to with transport efficiency, the term in not clearly defined.
- p. 8840, l. 14: "...model simulations of CO." AIRS wasn't used for the other tracers.
- 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.
- p. 8841, l. 21f: for the SO2/SO4 ratios the units are not correct

Technical Comments

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

- p. 8825, l. 7: explain the abbreviation "AIRS"
- p. 8826, Il. 12: "...transport of air from mid-latitudes to the Arctic..."
- p. 8827, l. 14: the abbreviation AIRS hasn't been fully explained yet in the text
- p. 8827, l. 27: "sources"
- p. 8828, I. 4: "has" instead of "have"
- p. 8828, l. 17: "using" instead of "paying careful attention to"
- p. 8828, l. 24: close parenthesis after link
- p. 8831, l. 1: "produced by the NASA..."
- p. 8831, II. 9-11: "three anthropogenic emission source regions from..., and two biomass burning emission source regions from..."
- p. 8831, l. 17: "identify"
- p. 8831, Il. 20 21: "panels"
- p. 8832, l. 18: "are shown in Fig. 3a..."
- p. 8833, l. 9, 21: "northern", and sometimes you use a capital letter for "Northern" sometimes you don't
- p. 8833, l. 12: "in the right column"
- p. 8833, l. 14: "biomass burning pollution contributions, "
- p. 8834, l. 2: "But contrary to the aerosol enhancement..."
- p. 8834, l. 5: "This observation..." instead of "interesting phenomenon"
- p. 8834, l. 15: "different air masses which were clean before..."
- p. 8834, l. 22: "with contribution from ..."

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- p. 8835, l. 15: "with only few percent of direct particulate sulfate emissions."
- p. 8835, I. 20: "over the US"
- p. 8835, I. 17: "The April campaign (ARCTAS-A) focused on the ..."
- 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.
- p. 8838, l. 2: "...confined to the lower and middle troposphere."
- p. 8838, l. 6ff: "... less efficient summer transport with 4
- 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..."
- p. 8838, l. 28ff: "both ASFF and BOBB in July which is in good agreement with the results from Fig.2a, b. " $\,$
- p. 8839, l. 2: what do you mean by "could", occasionally, has happened in a specific incident?...
- p. 8838, I. 7: "in April" instead of "from"
- p. 8839, I. 21: "Arctic" instead of "North Pole"
- p. 8839, l. 23: again, what do you mean with "could"?
- p. 8840, l. 5 "during ARCTAS-B"
- p. 8840, I. 8: "can" instead of "could"
- p. 8840, l. 24: "...(accounting for...)"
- 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.

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

Fig. 3 a, b the waypoint numbers are too small and sometimes can' 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.

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

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 8823, 2012.