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Interactive comment on “Multi-model study of chemical and physical controls on transport of anthropogenic and biomass burning pollution to the Arctic” by S. A. Monks et al.

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Interactive comment on “Multi-model study of chemical and physical controls on transport of anthropogenic and biomass burning pollution to the Arctic” by S. A. Monks et al. Response to Anonymous Referee #1

The authors examine the performance of eleven state-of-art models participating in the POLARCAT Model Inter-comparison Project over Arctic and lower latitude emission regions by evaluating their simulated key atmospheric components (CO, O₃, OH, and H₂O), using observations from aircraft, surface stations, and satellite. They further

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investigate the contributions of dynamic transport and OH chemistry to the diversity of simulated Arctic pollution using two designed tracers. This is an interesting and valuable study. The paper is well written. I recommend publishing the paper after the authors address the following minor comments.

The authors would like to that the reviewer for taking the time to read the manuscript and suggest changes. The suggestions have been taken on board and have helped produce a better manuscript. The comments are addressed individually below.

Specific Comments Page 25291 lines 14-20 and Page 25292 lines 1-13: It might be better to give a table summarizing the observed atmospheric components, the used technologies, and the measured accuracies from the aircraft campaigns of MOZAIC, ARCTAS-A, ARCTAS-B, POLARCAT-GRACE, and POLARCAT-France.

I have added a table to show all the observed data and species used along with measurement uncertainties. This has led me to change the text in Section 3 ('Observations') as follows. Added: 'The measurement uncertainties and techniques have been summarised in Table 1'

Removed, pg25291. L16-19: 'CO is measured by an infrared analyser, which is accurate to ± 5 ppbv (Nédélec et al., 2003), and O₃ is measured by a ultraviolet (UV) absorption instrument with an accuracy of ± 2 ppbv (Thouret et al., 1998).'

Removed, pg 25292, L3-4: 'OH was measured by laser induced fluorescence aboard this flight, which has relatively high measurement uncertainties (± 40 %) (Brune et al., 1999).'

Removed, pg 25292, L6-10: 'The POLARCAT-France ATR-42 measured CO using an infrared absorption analyser (Nédélec et al., 2003) and O₃ using an UV absorption instrument (Ancellet et al., 2009). The POLARCAT-GRACE Falcon also measured CO but used a vacuum UV fluorescence instrument (Gerbig et al., 1999) and O₃ was measured using a UV absorption analyser (Roiger et al., 2011).'

Added reference in new table: Sachse, G.W., and G.F. Hill, Fast-response, high precision carbon monoxide sensor using a tunable diode laser absorption technique, J. Geophys. Res., 92, 2071–2081, 1987.

Page 25293 – Page 25298: Reorganize sections 4.1, which addresses tracer seasonality, and 4.2, which addresses tracer vertical distribution. Aircraft measurements discussed in section 4.2 contain seasonal information, while satellite MOPITT data discussed in section 4.1.2 give CO vertical information.

The Section 4.1. and 4.2 subsection headers have been removed so the subsections (e.g. 4.1.1 & 4.1.2) now become subsections. This gets rid of any inconsistent grouping of comparisons.

Page 25294 lines 23-30: I am confused by the authors' explanation here for GEOSChem performance over Barrow. If including halogen chemistry is the reason for GEOS-Chem's better performance during March, why does this model still significantly underestimate O₃ year-around? Meanwhile, the authors indicate that GEOS-Chem model is also special in its accounting for transition metal catalyzed HO₂ uptake onto aerosol (page 25316 lines 1-5) and this new implementation might help GEOS-Chem performance during springtime. The paper would be more valuable if the authors could explore the contributions of these two chemical mechanisms more thoroughly.

It is agreed that the GEOS-Chem model is lower than observations throughout the year. However, it is only noticeably lower than the other models between March/April and July. Without separating out the different effects of HO₂ uptake and halogen chemistry it is hard to know what role these processes play in the negative model bias or whether there are other model deficiencies to blame. Whilst it would be valuable to have different model simulations from the GEOS-Chem GFDL group to investigate this further, unfortunately the group were not able to provide these simulations. This would definitely be an interesting point to pursue in the future.

The paragraph has been changed to address these comments:

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‘At Barrow, observations show a decrease in O₃ in March due to well-known halogen-induced ozone depletion events (ODE) in the boundary layer at this location (Barrie et al., 1988; Helmig et al., 2007). In contrast, most models simulate increasing concentrations between February and April, most likely due to the lack of halogen chemistry in the models, resulting in low correlations at Barrow. The GEOS-Chem model, which does include halogen chemistry, is still not able to capture the seasonal transition between February and April even though in March the absolute concentrations agree well. This model also simulates O₃ that is lower than the other models and observations between late-spring/early-summer, resulting in a larger negative bias against observations compared to other models. It is not clear what is causing this bias in GEOS-Chem, however, it occurs at the same time when simulated CO is higher than simulated by the other models suggesting it is related to the model chemistry, not transport.’

Page 25297 lines 15-26: The argument of vertical transport resulting in the vertical response of NMGE over North America is not convincing. If this hypothesis is true, we would expect a similar pattern of NMGE over Asia since vertical transport is typically more vigorous there. Meanwhile, the NMGE vertical pattern over North America is not supported by CO vertical profiles shown in Fig. 9.

The authors agree that if this hypothesis was true then we would expect to see the same distribution of RMSE over Asia and North America, suggesting there are other processes playing a role in the vertical distribution of the bias. I have rewritten the text as below to address this point.

‘The multi-model normalised mean gross error (NMGE, shown in Fig. 10), is found to show different vertical sensitivities depending on the region. Over North America, the NMGE is smaller at 700 hPa relative to 300 hPa, whilst over Europe the NMGE is lower at 300 hPa relative to 700 hPa. In contrast, the NMGE is similar at both 300 and 700 hPa over Asia. However, the overall range in error is relatively small in all these cases (10-14%). ‘

For the second point, whilst it is true that the MOZAIC data shows the largest bias over North America and Europe to be similarly placed in the lower free troposphere without any evidence of different vertical biases in the two regions, these comparisons are only done using data from the 18th June – 14th July in limited locations. The MOPITT NMGE have been calculated for the full yearly time series shown in Figure 6 and therefore this error is an estimate of error over the whole year at the certain altitudes (300hpa and 700hPa). For these reasons it is not unreasonable to get contrasting results.

This was mentioned briefly when comparing MOZAIC results to MOPITT later on page 25300: ‘The MOZAIC data shown here was collected at a limited number of locations, therefore the horizontal coverage is not as extensive as the MOPITT data and may explain some of the differences seen between these two observational datasets.’

Page 25300 lines 23-26: Why is the simulated CO by WRF-Chem 100 km larger than that by WRF-Chem 50 km?

The text has been changed to address this point in more detail as follows:

‘Global and regional models show similar overall performance, however the WRF-Chem 100 km resolution simulation has higher CO concentrations within the plume of enhanced CO at 900 hPa compared to the 50 km simulation, suggesting model resolution has important impacts on concentrations near emission sources. Whilst it is expected that a model run at a higher horizontal resolution would simulate higher concentrations, due to the emissions being subjected to less numerical dilution due to the smaller spatial resolution of a 50 km grid box, the model will also become more sensitive to emission location errors where emissions may be emitted into a neighbouring grid boxes. This will be particularly important for the model performance in the Arctic, where long-range transport of plumes plays an important role.’

Page 25302 line 23: Please clarify what “low model bias” means. Does it imply the model’s error is small or the model underestimates the observation?

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Changed from 'low model bias' to 'negative model bias'.

Page 25303 lines 10-12: This is an interesting issue that merits further discussion. It might indicate that some fundamental model processes, e.g. vertical convection, need to be improved.

The paragraph has been changed to the following to mention this point:

'For CO, models exhibit a negative bias over both source regions between 2 and 6 km (800–500 hPa). This bias can also be seen in some models in the MOPITT comparisons at 700 hPa in June and July (see bottom panel, Fig. 6). As the models show good agreement at the surface, this negative CO bias may be related to a problem with the simulated export of primary emissions from the boundary layer to the free troposphere suggesting that model transport processes, such as convection, need to be evaluated in more detail near source regions. This bias could also be a result of missing emissions in the inventories near the flight locations.'

Page 25308 line 16: Please add "and biomass burning" after "anthropogenic".
Changed to:

'The idealised tracers are used to compare the anthropogenic and biomass burning contributions from the three different source regions to the Arctic throughout the year. This is the first time biomass burning contributions have been summarised in this way. However, it is possible to compare the anthropogenic contributions found in this study to those in previous studies.'

Page 25317 line 1: Please add "Bian et al., 2013" before "Law et al., 2014".

Bian et al., (2013) is referenced in more detailed discussions throughout the paper (pg 25298, pg25300). In the conclusions I wanted to reference the POLARCAT overview paper of all the campaign related published results. Bian et al., (2013) is referenced within this paper. If I was to reference Bian et al., (2013) here then I would also need to reference many other relevant papers. And I have therefore changed '(Law et al.,

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2014)' to '(Law et al., 2014 and references therein)'.

Page 25317 lines 4-7: If over diffusion during long-range transport is an issue, it should apply to both O3 and CO coming from mid-latitude sources.

Yes, this is true, over diffusion of CO could be playing a role as evident by a negative bias in CO, which is mentioned on pg 25317, L3. I have added a sentence to the following paragraph to reflect this:

'O3 perturbations associated with the plumes also show large inter-model variability, which may point to different O3 production efficiencies in models. This could also be explained by deficiencies in model transport of mid-latitude sourced air masses over long distances into the Arctic in coarse global models, where plumes may become overly diffusive. This may also play a role in the negative bias in modelled CO within the same plumes.'

Figures 3: How is the value of RMSD from the figure supposed to be read?

The authors apologise, but there was a mistake in the caption. The RMSD is shown by the blue dotted lines (not black as stated). This has been corrected in the caption.

Technical corrections 1. Page 25298 line 9: Change DC8 to ARCTAS. 2. Changed from 'DC8' to ARCTAS-A (spring) and ARCTAS-B (summer) Page 25305 lines 6-7: Delete the sentence "Firstly, it is : : :". Deleted 'Firstly, it is'. Page 25306 line 11: Delete one of "the" in "the the". Deleted extra 'the' Page 25314 line 15: Delete "in the". Deleted 'in the' on line 16. Page 25316 line 4: Change "OF" to "of". Change to 'of' Figure 10: add "ATR = : : :." after "FAL = Falcon". Changed to 'POLARCAT_FAL=POLARCAT-GRACE, POLARCAT_ATR=POLARCAT-France'

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