

Interactive comment on "Transport of anthropogenic and biomass burning aerosols from Europe to the Arctic during spring 2008" by L. Marelle et al.

Anonymous Referee #2

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The authors apply WRF-CHEM and FLEXPART-WRF to a case study of airborne observations during April 9-11, 2008 as part of POLARCAT. Initially they compare simulated aerosol properties from WRF-CHEM with the airborne observations and with ground-based observations from the EMEP network to evaluate WRF-CHEM. They apply WRF-CHEM and FLEXPART-WRF to examine the origins of the aerosol measured from the aircraft and to estimate the direct radiative forcing impacts (at the TOA) of the aerosol split between European anthropogenic sources and biomass burning sources. In terms of forcings, the results are not very different from previous estimates, but the location of the measurements and the connections between the modelling and

C10393

observations offer a unique and useful perspective that I believe is relevant for ACP. The paper is well organized and written. A number of minor comments/questions follow:

1. Page 7, lines 1-2 – Why would you assume that SOA formation is low at high latitudes and in Europe but not sulphate formation? Both require oxidants, and gasphase NO3, which can be formed under low-light conditions, is a pretty good oxidant for many VOCs. You discuss that you model too much aerosol nitrate at the possible expense of organics. Perhaps there is some connection.

2. Page 11, line 18 to Page 12, line 5 – The discussion here is compromised by the fact that you have no SOA formation in your model. That said I tend to agree with your statement on lines 12-14 of page 12.

3. Page 13, line 2 - I suggest "baseline levels" in place of "background levels" here. Also, "clean Arctic background" is misleading. For example, at Alert, Nunavut, Canada 1 ug/m3 is more typical of polluted air masses. There is no basis for your reference to 'clean Arctic background', other than choosing the lowest values you measure. In the winter/spring, the Arctic baseline aerosol is elevated, and it is neither 'clean' nor 'background'.

4. Page 14, lines 12-16 – Your background aerosol in Fig 7 is sea salt, and that contributes the peak of 16 ug/m3 that you refer to on line 1 of page 13, where you also refer to background levels being about 1 ug/m3. The discussion on these points needs clarification.

5. Page 15, lines 10-15 – What do you mean by "large" particles? In all four cases, the model overestimates the number concentrations of particles in the bin that covers approximately 80 nm -150 nm. Since that size range includes the lower size of particles that typically activate in cloud (and therefore define the cloud droplet number concentrations), the model is over-predicting the CCN. The authors MAY be right in their statement that the aerosol optical properties will be correctly represented, but it is not clear from the comparisons in Figure 8 that that is true. If the authors are truly

concerned that the "aerosol impacts are treated accurately", then their size distribution comparisons should at least take the form of 1) a comparison of number concentrations greater than appropriate sizes in order to address the CCN issue, and 2) a comparison of the surface area distributions using a linear ordinate instead of a logarithmic ordinate.

6. Page 1, line 25 - had undergone significant ...?

7. Page 1, line 28 - here is it during 4-days and on line 22, it is during the campaign. Should be clarified.

8. Page 3, line 1 - By "low absorbing aerosols", do you mean aerosols with weak absorbing properties or do you mean absorbing aerosols low in the atmosphere?

9. Page 4, line 26 – are rather than "is".

10. Page 5, line 29 – define "background". I assume you mean air free from recent pollution sources as opposed to aerosol formed from only natural sources.

11. Page 8, lines 5-10 – Clarify on line 5 that the NODIRECT includes turning off the semi-direct as well. Also, somewhere earlier in the manuscript you should define direct and semi-direct.

12. Page 8, lines 12-17 – Could you not just calculate the backscatter ratio at 532 directly without the added step of scaling to the simulated Angstrom exponent?

13. Page 10, line 18 - replace "in which" with "when".

14. Page 11, line 12 - "... aerosols, enabling the relatively good PM2.5 agreement".

15. Page 14, line 31 – Size distribution is a fundamental microphysical property. Clarify "microphysical properties", by which I assume you mean CCN.

16. Page 16, line 11 – "in" rather than "on"; line 13 – I don't see where the black line in Fig 9 goes below -10 ug/m3, yet you say -12.

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17. Page 16, lines 11-26 – It is interesting that despite the stronger scavenging in the "K" case, the size distributions for 'K' (both modelled and observed) show considerably more particles larger than both 100 nm and 1um diameter. As you mention, the plume age may enhance the larger particles, but it seems that stronger wet scavenging should reduce the numbers of larger particles?

18. Page 17, lines 10-11 – the correlation is unclear, and during the time from about 11:10 to 11:40 the correlation does not appear to be as you describe it.

19. Page 17, lines 29-30 – Reasoning is needed here. If condensation processes are underestimated here, why not everywhere? Why does it have to be condensation, and not say reduced precursor emissions including possibly a lack of SOA? Address again in lines 9-10 on page 18.

20. Page 18, line 26 - "...features as were observed..."

21. Page 20-21 - Since there appears to have been a significant level of cloud cover, do you think there was potential for significant indirect effects associated with this aerosol?

22. Page 21, lines 20-26 – From your figure 12, the BC enhancement is relatively weak near the surface for the anthropogenic aerosol and even weaker still for the BB aerosol. So the statement that BC is "especially enhanced at the surface" seems to be out of line. What is the calculated level of surface cooling, and how might that compare with the heating potential from BC deposition?

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 28333, 2014.