

Answer to reviewer #2:

COMMENT: 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 gas phase 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.

RESPONSE: The first part of the comment has been addressed in the response to reviewer #1. We thank the reviewer for this insight on the possible link between nitrate overestimation and the lack of a SOA mechanism. This connection is now mentioned in section 4, where we discuss the overestimation of nitrate in WRF-Chem compared to EMEP measurements.

COMMENT: 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.

RESPONSE: We now highlight in this section that the underestimation of OC is likely associated with the lack of a SOA mechanism in our simulations.

COMMENT: 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’.

RESPONSE: We replaced “background levels” with “baseline levels” and “clean Arctic background” with “baseline aerosols”.

COMMENT: 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.

RESPONSE: In Figure 7 and associated discussion, “background” has been replaced by “unpolluted” and the meaning of this word (air free of recent pollution of sources) has been clarified.

**COMMENT: 5. Page 15, lines 10-15 –
What do you mean by “large” particles?**

RESPONSE: We replaced the term “large particles” by “particles larger than 200 nm”.

COMMENT: 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.

RESPONSE: We calculated the model normalized mean bias in terms of total number concentrations for particles larger than 80 nm. We obtain +57 %, +42 %, +98 % and +39 % for plumes I-J, K, M-N and L-O respectively. This overestimation means that the model will indeed probably overestimate CCN concentrations. However, we're not quantifying the aerosol indirect effect in this paper, so we leave further investigation of these issues for future studies. In order to address this comment, we have modified the section discussing aerosol size distributions from:

“Aerosol optical and microphysical properties are very sensitive to their size distributions (Boucher, 1998; Dusek et al., 2006). To ensure that aerosol impacts are treated accurately in the CTL simulation, modeled aerosol number size distributions are validated against in situ measurements for selected plumes.”

To (updated text):

“We evaluate model predictions of aerosol size distributions, which are known to be important for the optical properties (e.g. Boucher, 1998) presented later in the paper. Activation in clouds, which is outside the scope of the present study, is also sensitive to aerosol size distributions (Dusek et al., 1996).”

Finally, the most direct comparison with the measurements is using aerosol number concentrations, which are often shown on a log scale in order to display the full distribution, so Figure 8 has been left as is in the paper.

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

RESPONSE: This has been updated to: “... had undergone significant wet scavenging...”

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

RESPONSE: This text is confusing because the plumes sampled during the three POLARCAT-France flights were only present during 4 days in the Arctic (> 66.6 °N, not further north than 75 °N). The later part of the abstract refers to average results during the whole 4-day event, whereas the earlier part refers to results for plumes as they were sampled during the 3 POLARCAT-France flights. We updated the text from:

“Evaluating the regional impacts in the Arctic of this event in terms of aerosol vertical structure, we find that during the 4-day presence of these aerosols in the lower European Arctic (<75°N), biomass burning emissions...”

To (updated text):

“During this event, aerosols resided in the Arctic (> 66.6 °N) for 4 days. During this period, we find that biomass burning emissions...”

COMMENT: 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?

RESPONSE: “low absorbing aerosols” has been updated to “aerosols with weak absorbing properties”.

COMENT: 9. Page 4, line 26 – are rather than “is”.

RESPONSE: This has been updated to “However, clouds mostly impacted in-situ measurements at lower altitudes (< 2 km) and data are available for most periods of interest”

COMMENT: 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.

RESPONSE: This has been updated to: “Stations from the EMEP network are typically outside of urban centers and are intended to represent air free from local pollution sources.”

COMMENT: 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.

RESPONSE: This has been updated to: “(4) a simulation with the aerosol direct interaction with short wave radiation disabled, thus switching off the direct and semi-direct aerosol effects (NODIRECT)”. The direct and semi-direct effects have been defined in the introduction: “Aerosols play a key role in the climate system, through their absorption and scattering of solar radiation (direct effect, e.g. Haywood et Shine, 1995; Charlson et al., 1992), and through their impacts on cloud formation by modifying relative humidity and atmospheric stability (semi-direct effect, Ackerman et al., 2000) and by changing cloud properties and lifetime (indirect effect, Albrecht et al., 1989; Twomey, 1977).”

Albrecht, B. A.: Aerosols, Cloud Microphysics, and Fractional Cloudiness, Science, 245, 1227–1230, 1989.

COMMENT: 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?

RESPONSE: Backscatter ratios at 400 nm and 999 nm (used to calculate the Angström exponent) are default outputs in our version of WRF-Chem, and are calculated online. Backscatter ratios could be calculated at 532 nm directly by using a different setup of WRF-Chem or by modifying the radiation code, but new simulations would be required. Since the quantitative comparison of the LIDAR profiles and simulated results is not the main focus of this work, we decided to keep to the approach using the Angström exponent.

COMMENT: 13. Page 10, line 18 – replace “in which” with “when”.

RESPONSE: This has been updated to: “when high winds were observed”.

COMMENT: 14. Page 11, line 12 – “aerosols, enabling the relatively good PM2.5 agreement”.

RESPONSE: This has been updated to: “This suggests that the overestimation of NO_3^- and NH_4^+ might be compensated in terms of overall mass by an underestimation of organic carbon (OC) aerosols, resulting in relatively good $\text{PM}_{2.5}$ agreement.”

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

RESPONSE: This was addressed in the response to comment 5.

COMMENT: 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.

RESPONSE: This has been updated to: “The magnitude of wet scavenging along transport, also represented in Fig. 9C and 9D, is ...”

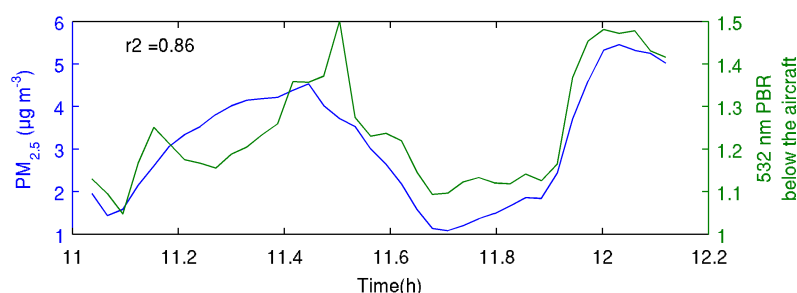
The typo in the next part has been corrected: “As expected, strong PM_{10} depletions, reaching -10 $\mu\text{g m}^{-3}$ (-74 %) are associated with precipitation during ...”

COMMENT: 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?

RESPONSE: The modeled “K” plume experienced stronger absolute wet scavenging ($-17 \mu\text{g m}^{-3}$ compared to $-10 \mu\text{g m}^{-3}$ for the anthropogenic plume) but lower relative wet scavenging (-55% compared to -74% for the anthropogenic plume), which, along with plume age, may explain why the K plume contains more particles larger in the $500 \text{ nm} - 2 \mu\text{m}$ diameter range. Plumes K and J were also sampled at different altitudes and did not originate from the same source, which could also play a role.

COMMENT: 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.

RESPONSE: Since this correlation was unclear in Figure 10A and 10B, we added a figure in the supplement (Figure S5, shown below) comparing the Pseudo Backscatter Ratio (PBR) at 532 nm just below the aircraft with $\text{PM}_{2.5}$ measured in-situ during this part of the 9 April 2008 flight. The correlation, while not perfect, is quite good ($r^2 = 0.86$). We included this value in the text.



COMMENT: 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.

RESPONSE: This has been updated to: “This means that the discrepancy in this layer probably corresponds to underestimated growth by condensation, which could be associated with underestimated precursor emissions including a lack of SOA. This is in agreement with the comparison of the modeled and observed size distributions of aerosols in mixed plumes, discussed in Sect 5.1, which indicated underestimated particle growth in the older mixed plumes.”

We also updated the text: “As discussed before, this confirms that the underestimation of $\text{PM}_{2.5}$ in this layer may be due to insufficient growth by condensation in this plume, possibly related to underestimated precursor emissions and a lack of SOA formation.”

COMMENT: 20. Page 18, line 26 – “features as were observed...”

RESPONSE: This has been updated to: “This average profile shows the same general features as were observed in-situ”

COMMENT: 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?

RESPONSE: During this transport event, clouds are often collocated with aerosol layers, meaning that cloud/aerosol interactions could be significant. We updated the part of the conclusion where we discuss aerosol indirect effects to mention this: “These radiative effect values do not include the impacts of cloud/aerosol interactions, which could be significant due to the extensive cloud cover in Northern Scandinavia during this transport event. This indirect effect could be quite large and compensate the warming effect of European aerosols over

snow and ice-covered surfaces. Moreover, the indirect aerosol effect is still uncertain, especially in the Arctic, and further work is needed to estimate its magnitude.”

COMMENT: 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?

RESPONSE: We updated the text to:

“At the surface, the direct aerosol effect causes local cooling for all types of land surfaces, including snow and ice (-1.1 W m^{-2} DSRE on average, -2.75 W m^{-2} at noon over Scandinavia and Finland). However, we also show in Figure 12 that BC was enhanced at the surface in anthropogenic plumes, which could lead to surface warming through the effects of BC deposited on snow. Black carbon deposition is not coupled to snow albedo in WRF-Chem 3.5.1, however the global model study of Wang et al. (2011) showed that in spring 2008 (April–May), significant levels of anthropogenic BC (1 to $5 \text{ mgC m}^{-2} \text{ month}^{-1}$) were deposited on snow in Northern Europe, leading to 1 to 2 % change in the regional albedo of snow and ice. This change in snow albedo was estimated to cause a radiative effect of 1.7 W m^{-2} in April-May (average value for the Arctic $> 60^\circ \text{ N}$). Wang et al. (2011) do not show the geographical distribution of this forcing, which should be higher in Scandinavia and Finland because the snow-albedo change from BC deposition is higher in their study in continental Eurasia than in the rest of the Arctic.”