

Answer to reviewer #1:

COMMENT: p. 28334, last line: Is Finland not part of Scandinavia?

RESPONSE: Finland is not traditionally considered to be part of Scandinavia (Norway, Denmark and Sweden). The reunion of Finland and Scandinavia is sometimes referred to as Fennoscandia, but we left “Scandinavia and Finland” in the text.

COMMENT: p. 28336, l. 1: “... chemical-transport models.” This needs a reference, e.g., Shindell et al. (2008)

RESPONSE: We added the reference to Shindell et al. (2008)

Shindell, D. T., Chin, M., Dentener, F., Doherty, R. M., Faluvegi, G., Fiore, A. M., Hess, P., Koch, D. M., MacKenzie, I. A., Sanderson, M. G., Schultz, M. G., Schulz, M., Stevenson, D. S., Teich, H., Textor, C., Wild, O., Bergmann, D. J., Bey, I., Bian, H., Cuvelier, C., Duncan, B. N., Folberth, G., Horowitz, L. W., Jonson, J., Kaminski, J. W., Marmer, E., Park, R., Pringle, K. J., Schroeder, S., Szopa, S., Takemura, T., Zeng, G., Keating, T. J., and Zuber, A.: A multi-model assessment of pollution transport to the Arctic, *Atmos. Chem. Phys.*, 8, 5353-5372, doi:10.5194/acp-8-5353-2008, 2008.

COMMENT: section 2.1.: please add information on the time resolution of the measurements

RESPONSE: We added information about the time resolution of the meteorological data (1 s), the GRIMM optical counter (1 s) and the SMPS particle sizer (140 s).

COMMENT: p. 28338, l. 1: please include information on the overall number of data points to show that the following analysis is statistically relevant.

RESPONSE: The number given in the text (88 %) was based on values for the GRIMM OPC (22,013 data points for the 3 flights). For the SMPS particle sizer, the coverage is 98 % (158 data points for the 3 flights). We added this distinction between SMPS and GRIMM in the text, as well as the number of data points for each instrument.

COMMENT: p. 28338, ll. 2 – 7: Where does this information come from? Either provide a reference or give a concrete example based on the data you use.

RESPONSE: The numbers in “94 % of the measured 20 nm to 2 µm mass distribution is located in the lower size range 20nm to 1.6 µm” were determined using all available POLARCAT-France spring data during the 9, 10 and 11 April flights. We updated the text to: “The contribution of particles in the 2–2.5 µm diameter range to PM_{2.5} is missing from this estimation. However, we determine it is negligible, because 94 % of the measured 20 nm to 2 µm mass distribution in the POLARCAT-France dataset is located in the lower size range 20 nm to 1.6 µm...”

COMMENT: p. 28339, l. 10: What type of data, mass concentration, chemical composition, etc.? Please specify.

RESPONSE: We use EMEP PM_{2.5} mass concentration, and chemical composition in SO₄²⁻, OC, BC, NH₄⁺, NO₃⁻ from filter measurements. The text has been updated to include this information.

COMMENT: p. 28340, l. 8: Please provide more specifics on what “aerosol/cloud interactions” includes.

RESPONSE: The text has been rewritten to be more specific about aerosol/cloud interactions

implemented in WRF-Chem/MOSAIC. Specifically, we have added a description of how MOSAIC represents the first and second aerosol indirect effects.

“MOSAIC aerosol processes include nucleation, evaporation, coagulation, condensation, dry deposition, and aerosol/cloud interactions, including aerosol activation as cloud condensation nuclei (CCN), cloud chemistry, and within and below cloud wet scavenging. Eight bins represent the size distribution of each aerosol species between 39 nm and 10 μm . Interstitial and cloud-borne aerosol particles are treated explicitly, and modeled aerosols can be activated or re-suspended depending on saturation, particle size and aerosol composition. Aerosol activation changes cloud droplet number concentrations in the Morrison microphysics scheme, which is coupled with the Goddard shortwave radiative scheme (first indirect effect). Aerosol activation also affects cloud lifetime by influencing precipitation (second indirect effect). Aqueous chemistry in clouds is based on Fahey and Pandis (2001), and includes oxidation of S(IV) by H_2O_2 , O_3 , and other radicals, as well as non-reactive uptake of NH_3 , HNO_3 , HCl , and other trace gases.”

COMMENT: p. 28340, l. 17 – 19: “... and SOA formation is likely to be low in Europe: This needs a reference.

RESPONSE: We included a reference to Bessagnet et al. (2008), indicating that 75-95% of SOA in Europe were associated with biogenic sources in 2003, and a reference by Karl et al. (2009), showing that biogenic VOC emissions are relatively low in Europe in March and April. Bessagnet et al. (2008) also point out that on average, SOA concentrations are much lower in northern Europe than in southern Europe. However Frossard et al. (2011) also determined that during the POLARCAT-France flights (April 2008), SOA formation contributed to the organic fraction of aerosols measured in the Scandinavian marine boundary layer. We included this discussion in the text, and mentioned more clearly that the modeled organic matter is likely too low because of this lack of SOA.

Bessagnet, B., Menut, L., Curci, G., Hodzic, A., Guillaume, B., Liousse, C., Moukhtar, S., Pun, B., Seigneur, C., and Schulz, M.: Regional modeling of carbonaceous aerosols over Europe-focus on secondary organic aerosols, *J. Atmos. Chem.*, 61, 175–202, doi:10.1007/s10874-009-9129-2, 2008.

Frossard, A. A., Shaw, P., Russell, L. M., Kroll, J. H., Canagaratna, M. J., Worsnop, D. R., Quinn, P. K., and Bates, T. S.: Springtime Arctic haze contributions of submicron organic particles from European and Asian combustion sources, *J. Geophys. Res.*, 116, D05205, doi:10.1029/2010JD015178, 2011.

Karl, M., Guenther, A., Köble, R., Leip, A., and Seufert, G.: A new European plant-specific emission inventory of biogenic volatile organic compounds for use in atmospheric transport models, *Biogeosciences*, 6, 1059–1087, doi:10.5194/bg-6-1059-2009, 2009.

COMMENT: p. 28341, l. 8: Not clear to what these numbers refer. In-domain, global, other?

RESPONSE: The numbers refer to in-domain HTAPv2 anthropogenic emissions. This is now specified in the text.

COMMENT: p. 28341, l. 27 – 29: How exactly do you make this estimation?

RESPONSE: This estimation is made by comparing along the flight track $\text{PM}_{2.5}$ from the NOANTHRO and NOFIRES simulations with $\text{PM}_{2.5}$ from the CTL simulation. It is described in more detail in section 5.1. The text has been updated to make it clearer where in the paper

each of these simulations are used.

COMMENT: p. 28345, l. 16 – 18: What exactly do you mean by “compensated”? Does this refer to the overall mass? Or to other characteristics such as hygroscopicity, optical properties, size, shape? Be more specific.

RESPONSE: In this case, “compensated” refers to the overall mass. The influence on optical properties and hygroscopicity is discussed in the end of section 4. The text has been updated: “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: p. 28345 f, l. 25 – l. 3: How do Tuccella et al. (2012) explain the deficiencies in modeling OC?

RESPONSE: Tuccella et al. (2012) state that this deficiency in OC is due to the incomplete description of SOA formation in their mechanism, including the lack of oxidation of biogenic monoterpenes and a “limited treatment of anthropogenic VOC oxidation”. The text has been rewritten to include these details.

COMMENT: p. 28346, l. 19: What is the number in brackets? Do you mean kappa 0.14? Specify.

RESPONSE: This has been updated to: “The bulk hygroscopicity of OC ($\kappa = 0.14$) is lower than the one of NO_3^- and NH_4^+ ($\kappa = 0.5$) in MOSAIC. »

COMMENT: p. 28346, l. 25: Do you mean all radiative effects or only the direct? Be more specific.

RESPONSE: This has been updated to “... to investigate the direct and semi-direct radiative effects”.

COMMENT: p. 28347, l. 26: Give a reason why you use 20 %.

RESPONSE: In this section and the rest of the paper, we aim to highlight the difference between air masses significantly influenced by biomass burning (BB) and air masses mostly influenced by anthropogenic emissions. We used a threshold of 20 % to exclude other air masses weakly influenced (5 to 15 %) by BB on the 10 and 11 April flights (as seen on figure S2) and to identify air masses significant influenced by BB, up to 30-40%. However, this means that anthropogenic air masses sampled on 10 and 11 April are also somewhat influenced by BB (3% of $\text{PM}_{2.5}$ on average on the 9th, 6 % on the 10th, 7 % on the 11th). We used the same threshold of 20 % for anthropogenic plumes for consistency. The text has been updated to reflect this discussion.

COMMENT: p. 28349, l. 7: Include a reference for the underestimation by many global models.

RESPONSE: This was a mistake; see the paper of Schwarz et al., 2010, showing that global models often overestimate BC concentrations aloft in the Arctic. We thank the reviewer for pointing this out. The sentence has been updated to: “Plumes coming from the northern domain boundary, which are not studied in detail here, reflect the aerosols present in the MOZART 4 simulation used as the boundary conditions and point to a general underestimation.”

COMMENT: p. 28349, ll. 9 – 13: How does this composition compare to other in-situ measurements of pollution plumes in the Arctic that were measured during the several

POLARCAT (including ARCTAS and ARCPAC) campaigns? Is it special or similar to what we know already?

RESPONSE: Other in-situ measurements during POLARCAT generally indicate less nitrate and more organic matter in Arctic aerosols. For example, Brock et al. (2011) found 78 % OM and 20 % NO_3^- in biomass burning plumes during ARCPAC (32 % and 1 % for anthropogenic plumes). During ICEALOT, at the same time and location as the POLARCAT-France measurements, Frossard et al. (2011) found (excluding sea salt and black carbon) 30 % organic matter, 60 % sulfate and 1 % nitrate in the Scandinavian marine boundary layer. Airborne AMS measurements in the summer in Greenland during POLARCAT-France (Schmale et al., 2011) also indicate very low nitrate (below the detection limit) and high organic matter (50 to 90 %) in polluted plumes. This comparison also indicates that in our simulations, nitrate aerosols might have been formed at the expense of secondary organic aerosols due to the lack of a SOA mechanism. This discussion has been included in the text.

Schmale, J., Schneider, J., Ancellet, G., Quennehen, B., Stohl, A., Sodemann, H., Burkhardt, J. F., Hamburger, T., Arnold, S. R., Schwarzenboeck, A., Borrmann, S., and Law, K. S.: Source identification and airborne chemical characterisation of aerosol pollution from long-range transport over Greenland during POLARCAT summer campaign 2008, *Atmos. Chem. Phys.*, 11, 10097-10123, doi:10.5194/acp-11-10097-2011, 2011.

COMMENT: p. 28349, ll. 20 – 29: You elaborate on the deficiencies in representing nitrate, ammonium and OC with the model and then compare your results to Brock et al. (2011) for BC only. Include a discussion on the other chemical components as well.

RESPONSE: We now mention in this part of the text that aerosols measured by Brock et al. contain proportionally less sulfate and nitrate and more organic matter. This is, in part, due to the fact that they targeted biomass burning plumes richer in OC. However, as discussed earlier, the refraction indices for these different components are similar in WRF-Chem, while different black carbon concentrations are more likely to strongly change the magnitude of the aerosol direct and semi-direct effect.

COMMENT: p. 28350, l. 22: What is the diameter range of the accumulation mode you are referring to? Also specify the type of diameter.

RESPONSE: We now mention in the text, according to Quennehen et al. (2012), that the diameter ranges (Stokes diameter) for these accumulation modes are 90 – 500 nm for the anthropogenic plume, and 110 - 700 nm for the fire plume.

COMMENT: p. 28355, l. 20: What do you mean by should? Do they contribute or do they not?

Previous studies (e.g. Flanner et al., 2013) show that absorbing aerosols do contribute to Arctic warming in spring; this is the focus of the section following this statement. We have updated the text to: “Because the transport of pollution from Europe to the Arctic is especially efficient in late winter and early spring when the Scandinavian snow cover is still extensive, aerosols transported to the Scandinavian Arctic may contribute to enhanced local atmospheric heating rates in this region (Flanner et al., 2013). We investigate this by calculating the direct and semi-direct shortwave (0.125 to 10 μm wavelengths) radiative effect (DSRE) of aerosols at the Top Of Atmosphere (TOA), in regions significantly influenced by in-domain anthropogenic and biomass burning emissions. The DSRE, shown in Figure 13a, is estimated by taking the difference between the upward short wave TOA...”

COMMENT: p. 28356, l. 19 and following: How comparable are the values? Have the

plumes on average the same age, are the sources and characteristics comparable?

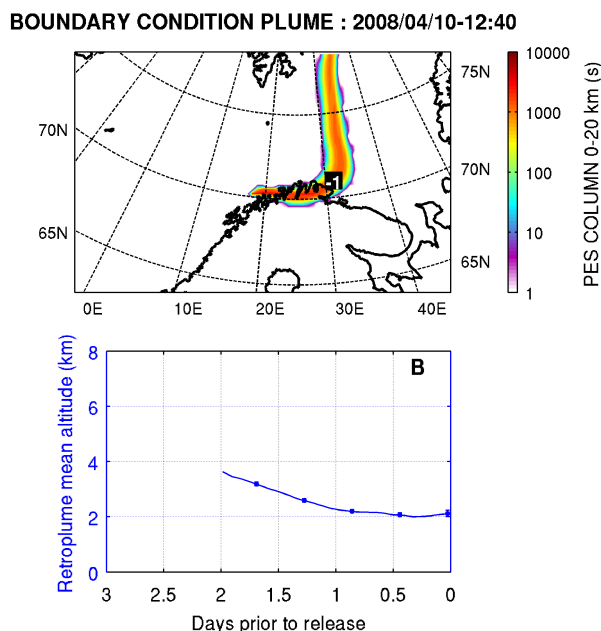
In this study, we focus on the springtime European Arctic and put our results into the context of other studies focusing on the same period, but in different locations. We summarize the other studies for comparison, but it is difficult to draw broader conclusions about whether they are representative of wider spatial and or temporal scales. To clarify this we added a sentence at the beginning of this section.

COMMENT: Figures S2: Describe what the letters mean in the plot.

RESPONSE: The caption has been updated to mention that: “Letter labels indicate anthropogenic (I, J, M, N) and mixed anthropogenic/fire (K, L, O) plumes investigated further.”

COMMENT: Figure S3a: The column integrated PES is hardly visible. I suggest to zoom into the region.

RESPONSE: We replaced Figure S3 with a zoomed-in version, shown below.



Technical comments

COMMENT: p. 28334, l. 7: Split the sentence, it is too long.

RESPONSE: This has been updated to: “Modeled PM_{2.5} is evaluated using EMEP measurements in source regions and POLARCAT aircraft measurements in the Scandinavian Arctic. Total PM_{2.5} agrees well with the measurements, although the model overestimates nitrate and underestimates organic carbon in source regions.”

COMMENT : p. 28336, l. 18: missing word “These studies pointed towards the needs...”

RESPONSE: This has been updated to: “These studies pointed out the need for modeling in order to quantify the influence of different processes and sources on aerosols observed during the campaign.”

COMMENT: p. 28340, l. 7: there is a closing bracket too much after “version Z”.

RESPONSE: This has been updated to: “(Carbon Bond Mechanism, version Z, Zaveri and

Peters, 1999)”

COMMENT: p. 28344, l. 5: delete “by”

RESPONSE: This has been updated to: “...and has been identified as a mixed anthropogenic and biomass burning plume originating from northeast Asia.”

COMMENT: p. 28344, l. 20: Under these conditions the experience might have been like a “fight” but you probably mean “flight”.

RESPONSE: This has been updated to: “over the Norwegian Sea during this portion of the flight do not impact”

COMMENT: p. 28346, l. 15: delete “of” in “...NH₃ could cause of an enhanced ...”

RESPONSE: This has been updated to: “overestimation of NH₃ could cause an enhanced formation of ammonium nitrate”

COMMENT: p. 28346, l. 22: The chemical formula for ammonium sulfate is incorrect.

RESPONSE: This has been updated to: “(NH₄)₂SO₄”

COMMENT: p. 28351, l. 8: There is on “large” too much.

RESPONSE: This has been updated to: “mixed plume is 3 to 5 days old and under the influence of emissions in a large region”

COMMENT: p. 28351, l. 15: Flanner (2013) would be another appropriate reference.

RESPONSE: It seems like the reference to Flanner et al. (2013) was meant to be integrated p. 28355, l. 15 instead of p. 28351, l. 15. We included this reference in the text: “Because the transport of pollution from Europe to the Arctic is especially efficient in late winter and early spring when the Scandinavian snow cover is still extensive, aerosols transported to the Scandinavian Arctic may contribute to enhanced local atmospheric heating rates in this region (Flanner et al., 2013).”

Flanner, M. G.: Arctic climate sensitivity to local black carbon, *J. Geophys. Res.*, doi:10.1002/jgrd.50176, 2013.

COMMENT: p. 28353, l. 4, a “,” is missing after “(Fig. 10c)”

RESPONSE: This has been updated to: “The PBR at 532 nm is compared to cross-sections of the simulated backscatter ratio (Fig. 10c), simulated PM_{2.5} (Fig. 10d) and ...”

COMMENT: p. 28353, l. 12: “ASPR” has not been introduced yet

RESPONSE: This was a mistake; this part has been updated to say “PBR” (Pseudo Backscatter Ratio).

COMMENT: p. 28354, l. 15: A “.” is missing between the sentences.

RESPONSE: This has been updated to: “...extent and vertical structure of the plumes. We now investigate the regional impacts of...”