

Dear Radovan Krejci

Thank you for your efforts to help with the editorial process with our manuscript. We have organized our response as follows. First, we include the responses to the referees uploaded on ACPD as part of the interactive discussion. Then, we include a track changes version of the manuscript with all removed text in red and new text in blue. The goal was to answer each reviewer comment and integrate the related changes into the revised manuscript (attached). We note that there are some small differences in wording in the revised manuscript compared to the online responses, which have been modified primarily for readability. Thanks in advance for your continued work as editor of this manuscript and I look forward to hearing from you in the coming weeks.

Best regards,

Louis Marelle

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 $\text{PM}_{2.5}$ 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 $\text{PM}_{2.5}$ mass concentration, and chemical composition in SO_4^- , OC, BC, NH_4^+ , NO_3^- 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., Burkhart, 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?

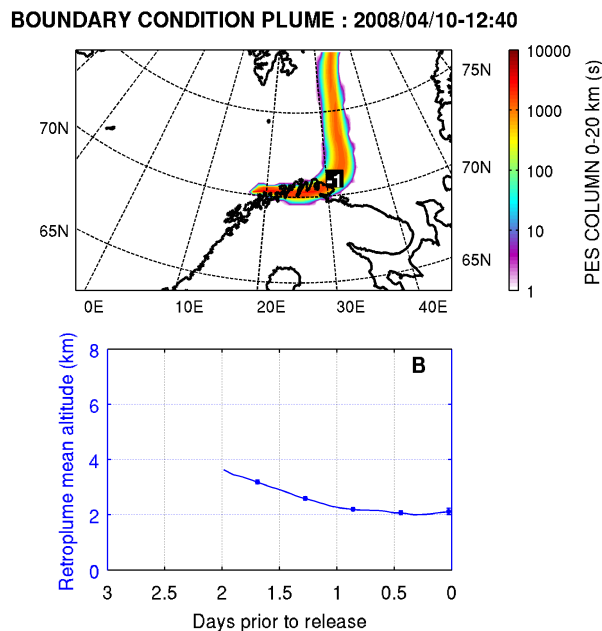
In this study, we focus on the springtime European Arctic and put our results into the context of other studies focusing on the same period, but in different locations. We summarize the other studies for comparison, but it is difficult to draw broader conclusions about whether they are representative 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...”

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 ...”

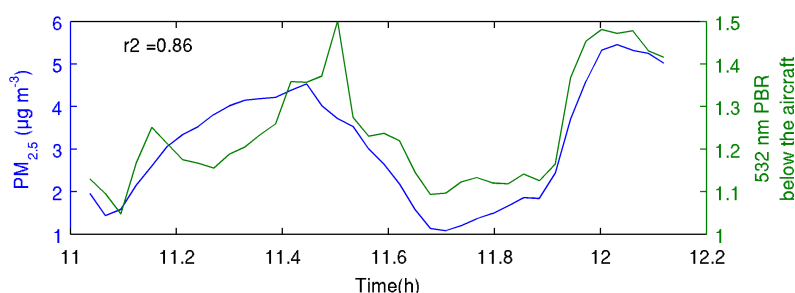
The typo in the next part has been corrected: “As expected, strong PM_{10} depletions, reaching -10 $\mu\text{g m}^{-3}$ (-74 %) are associated with precipitation during ...”

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.”

Transport of anthropogenic and biomass burning aerosols from Europe to the Arctic during spring 2008

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Abstract

During the POLARCAT-France airborne campaign in April 2008, pollution originating from anthropogenic and biomass burning emissions was measured in the European Arctic. We compare these aircraft measurements with simulations using the WRF-Chem model to investigate model representation of aerosols transported from Europe to the Arctic. Modeled $\text{PM}_{2.5}$ is evaluated using EMEP measurements in source regions and POLARCAT aircraft measurements in the Scandinavian Arctic, ~~showing a good agreement,~~ Total $\text{PM}_{2.5}$ agrees well with the measurements, although the model overestimates nitrate and underestimates organic carbon in source regions. Using WRF-Chem in combination with the Lagrangian model FLEXPART-WRF, we find that during the campaign the research aircraft sampled two different types of European plumes: mixed anthropogenic and fire plumes from eastern Europe and Russia transported below 2 km, and anthropogenic plumes from central Europe uplifted by warm conveyor belt circulations to 5–6 km. Both modeled plume types had undergone significant wet scavenging ($> 50\%$ PM_{10}) during transport. Modeled aerosol vertical distributions and optical properties below the aircraft are evaluated in the Arctic using airborne LIDAR measurements. ~~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 ($< 75^\circ\text{N}$)~~ Model results show that the pollution event transported aerosols into the Arctic ($> 66.6^\circ\text{N}$) for a 4-day period. During this 4-day period, biomass burning emissions have the strongest influence on concentrations between 2.5 and 3 km altitudes, while European anthropogenic emissions influence aerosols at both lower (~ 1.5 km) and higher altitudes (~ 4.5 km). As a proportion of $\text{PM}_{2.5}$, modeled black carbon and SO_4^{2-} concentrations are more enhanced near the surface. The European plumes sampled during POLARCAT-France were transported over the region of springtime snow cover in Northern Scandinavia, where they had a significant local atmospheric warming effect. We find that, during this transport event, the average modeled top of atmosphere (TOA) shortwave direct and semi-direct radiative effect (DSRE) north of 60°N over snow and ice-

covered surfaces reaches $+0.58 \text{ W m}^{-2}$, peaking at $+3.3 \text{ W m}^{-2}$ at noon over Scandinavia and Finland.

1 Introduction

Arctic haze, which is present during winter and spring, is a well known phenomenon that includes elevated concentrations of anthropogenic aerosols transported to the Arctic region (e.g. Rahn et al., 1977; Quinn et al., 2007). It was identified for the first time in the 1950s, when pilots experienced reduced visibility in the springtime North American Arctic (Greenaway, 1950; Mitchell, 1957). Further analysis showed that Arctic haze aerosols are mostly composed of sulfate, as well as organic matter, nitrate, sea salt, and black carbon (e.g. Quinn et al., 2002). Since local Arctic emissions are rather low, most air pollutants in the Arctic originate from transport from the mid-latitudes (Barrie, 1986). In late winter and early spring, Eurasian emissions can be efficiently transported at low-level in the Arctic (Rahn, 1981), while removal processes are particularly slow (Shaw, 1995; Garrett et al., 2011), causing elevated pollution concentrations in the lower troposphere. Surface aerosol concentrations in the Arctic are mostly influenced by European and West Asian emissions, while East Asian emissions have a larger influence in the upper troposphere (Fisher et al., 2011). Eurasian biomass burning emissions are thought to be major sources of Arctic pollution (Stohl, 2006; Warneke et al., 2010), but the magnitude of this contribution is still uncertain.

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, lifetime and precipitation \(indirect effects\)](#), Twomey, 1977; [Albrecht et al., 1989](#)). In the Arctic, several processes enhance the radiative impact of aerosols, including soot deposition on snow (Flanner et al., 2007), increased long wave emissivity in clouds in polluted conditions (Garret and Zhao, 2006), and the increased atmospheric heating effect of **low**

~~absorbing aerosols~~ aerosols with weak absorbing properties over snow or ice covered surfaces (Pueschel and Kinne, 1995; Haywood and Shine, 1995). Modeling studies by Shindell and Faluvegi (2009) and Jacobson, (2010) suggest that a good representation of aerosol composition and optical properties is critical to understand the Arctic energy budget. However, it is well known that aerosols amounts and properties in the Arctic are not well represented in global chemical-transport models - (Shindell et al. (2008)). For example, Schwarz et al. (2010) showed that black carbon in global simulations does not agree well with observations in the Arctic and varies greatly between models. This discrepancy, especially at high altitudes, may be caused, in part, by insufficient rainout (e.g. Wang et al., 2013).

To improve our understanding about air pollution in the Arctic, several airborne campaigns were conducted in the Arctic region during the International Polar Year in 2008 in the framework of POLARCAT (POLar study using Aircraft, Remote sensing, surface measurements and models, of Climate, chemistry, Aerosols, and Transport, see Law et al., 2014). As part of the international project POLARCAT, the POLARCAT-France spring campaign took place from 30 March to 14 April 2008, based in Kiruna, Sweden (67.8° N, 20.2° E). This campaign focused on Arctic cloud-aerosol interactions, satellite measurement validation, and transport of pollution plumes from mid-latitudes to the Arctic. During the campaign, several anthropogenic and biomass burning plumes originating in Europe and Asia were transported to the flight area and sampled during flights in April 2008 (Adam de Villiers et al., 2010; Quennehen et al., 2012). Adam de Villiers et al. (2010) analyzed the optical properties of aerosol plumes measured by airborne and spaceborne LIDAR, and Quennehen et al. (2012) studied aerosol ageing from size distributions measured in situ during POLARCAT-France spring. These studies pointed out the need for modeling to quantify the influence of different processes and sources on aerosols observed during the campaign.

The present study aims to improve our understanding about aerosol originating from Europe. In particular, we investigate the role of anthropogenic and biomass burning sources, transport pathways, aerosol ageing, and processes controlling the vertical distribution of aerosol plumes transported to the European Arctic in spring, and how they impact the aerosol burden and the aerosol radiative effect in this region. To achieve this objective,

measurements from the POLARCAT-France airborne campaign in the Scandinavian Arctic in April 2008 are analyzed in combination with simulations using the regional WRF-Chem model to investigate cases of aerosol transport from Europe to the Arctic. In Sect. 2, we describe the methods used in our study, including a description of the POLARCAT-France spring airborne aerosols measurements, and the EMEP ground based aerosol measurements used to validate the model over European source regions. Section 2 also includes an overview of the modeling tools employed, WRF-Chem and FLEXPART-WRF, and describes the simulations performed in this study. In Sect. 3, we present the synoptic scale meteorological situation over Europe during the campaign, and how this situation impacted long-range aerosol transport from Europe to the Arctic. In Sect. 4, the performance of the WRF-Chem simulation is evaluated using POLARCAT-France spring meteorological measurements and ground based aerosol measurements in source regions. In Sect. 5, modeled aerosol physical and optical properties are compared to POLARCAT-France spring airborne in situ and LIDAR measurements. We also investigate in Sect. 5 the sources of aerosols observed during the campaign. The results are used in Sect. 6 to evaluate the regional impact of this transport event in terms of aerosols burden and direct radiative effects.

2 Methods

2.1 POLARCAT-France spring campaign airborne measurements

During POLARCAT-France, the French ATR-42 research aircraft payload included two instruments to measure the particle size distribution: a Scanning Mobility Particle Sizer (SMPS, size range 20 to 467 nm, 88 channels, [140 s resolution](#)) and a GRIMM Optical Particle Counter (OPC, size range 0.1 to 2 μm , 8 channels, [1 s resolution](#)). For the full size distributions (20 nm to 2 μm), data from the two instruments are combined as described in Quennehen et al. (2012). The ATR-42 was equipped with a Counterflow Virtual Impactor (CVI) inlet (Schwarzenboeck et al., 2000) to sample aerosol particles and cloud droplets. In clouds, the CVI inlet was activated to remove interstitial aerosols and study cloud droplets

only. Therefore, aerosol size distributions are only available out of clouds. However, clouds mostly impacted in-situ measurements at lower altitudes (< 2 km) and data ~~is~~are available for most periods of interest for modeling long-range transport of aerosols to the region (SMPS: 158 data points, 98 % coverage above 1.5 km; GRIMM: 22,013 data points, 88 % data coverage above 1.5 km). $\text{PM}_{2.5}$ concentrations along the flight track are estimated by integrating the size distributions (20 nm to 2 μm), assuming that all particles are spherical and have a density of 1700 kg m^{-3} (Quennehen et al., 2011). The contribution of particles in the 2–2.5 μm diameter range to $\text{PM}_{2.5}$ 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 , and because large particles are unlikely to be transported over long distances.

During the campaign, airborne aerosol LIDAR profiles were measured below or above the aircraft by the LNG instrument (LIDAR LEANDRE Nouvelle Génération) (Flamant and Pelon, 1996; Adam de Villiers et al., 2010; Ancellet, 2014). Specifically, the LNG instrument measured aerosol optical properties at two wavelengths (532 and 1064 nm) providing information about the location of aerosol layers vertically (in our case below the aircraft). The vertical resolution of the data presented is 30 m (4 point average) and the horizontal resolution is 450 m (average of 100 LIDAR shots). In this work, we use the LNG measurements to study the spatial structure of aerosol layers below the aircraft and to analyze the representation of these aerosol layers in regional chemical transport modeling. For this purpose, we use the LNG measurements to calculate the pseudo backscatter ratio (PBR), defined as the ratio of the measured LIDAR total attenuated backscatter (including Rayleigh and aerosol contributions) to simulated molecular backscatter at a certain wavelength. The uncertainty for this ratio is estimated to be 10 % for the 532 nm channel and 20 % for the 1064 nm channel by Adam de Villiers et al. (2010). For this reason, we only use the 532 nm PBR in this study. In moderately polluted conditions (as observed during POLARCAT-France spring campaign), the PBR is close to the true backscatter ratio, defined as $R_T = \frac{(\beta_A + \beta_M)}{\beta_M}$, where β_A is the aerosol backscatter coefficient and β_M is the molecular backscatter coefficient, noting that the true backscatter ratio is equal to 1 in clear sky conditions, and is greater

than 1 in aerosol layers. Several aerosol plumes were sampled in situ and measured by LIDAR during three flights on 9, 10 and 11 April 2008. The associated flight tracks, over northern Norway and the Norwegian Sea/Barents Sea region, are represented in Fig. 1.

2.2 EMEP ground based measurements

The European Monitoring and Evaluation Programme (EMEP) network of ground-based measurements includes both aerosol $PM_{2.5}$ mass and aerosol chemical composition (available online from the EMEP database – <http://www.nilu.no/projects/coc/>). Stations from the EMEP network are typically outside of urban centers and are intended to represent ~~background and/or aged aerosol properties, as they are not co-located with large emissions~~ air free from recent pollution sources. We use the EMEP measurements of $PM_{2.5}$, as well as chemical composition in $SO_4^{=}$, OC, BC, NH_4^+ and NO_3^- to evaluate model aerosols from 1 April to 11 April -2008, using data from stations with either daily or hourly data. ~~In addition, stations~~ Stations are excluded if they have less than 75% data coverage during this period, and OC or BC measurements are excluded because of the lack of spatial coverage of measurements (4 stations for BC, 5 for OC). The locations of stations used for model comparison are shown in Fig. 1, including stations that measure $PM_{2.5}$ (33 stations) and stations that measure aerosol mass of $SO_4^{=}$, NH_4^+ , and NO_3^- (34, 31, and 28 stations respectively). The average data coverage for selected stations is 98%.

2.3 Model calculations: WRF-Chem and FLEXPART-WRF

2.3.1 WRF-Chem

Regional chemical transport model simulations are performed with the version 3.5.1 of the WRF-Chem (Weather Research and Forecasting, including Chemistry) model to provide further insight into the POLARCAT-France spring aerosol measurements. WRF-Chem is a fully coupled, online meteorological and chemical-transport mesoscale model (Grell et al., 2005; Fast et al., 2006). It has been successfully used in previous studies focused on the Arctic region (Sessions et al., 2011; Thomas et al., 2013) and to analyze airborne

aerosols measurements (e.g. Fast et al., 2012). The model setup including the representation of the planetary boundary layer, surface, radiative properties, convection, microphysics, gas phase chemistry, and aerosols is shown in Table 1. Specifically, gas-phase reactions were simulated with the CBM-Z mechanism (Carbon Bond Mechanism, version Z) (Zaveri and Peters, 1999) and aerosols are represented using the 8 bin sectional aerosol model MOSAIC (Model for Simulating Aerosol Interactions and Chemistry, Zaveri et al., 2008). MOSAIC aerosol processes include nucleation, evaporation, coagulation, condensation, ~~cloud chemistry, dry deposition, and~~ aerosol/cloud interactions, ~~dry deposition, including aerosol activation as cloud condensation nuclei, 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 ~~resuspended~~ ~~re-suspended~~ depending on saturation, particle ~~sizes~~ ~~size~~ and aerosol composition. ~~Aerosol activation changes cloud droplet number concentrations in the Morrison microphysics scheme, which is linked with the Goddard shortwave radiative scheme. Aerosol activation also affects cloud lifetime by influencing precipitation.~~ 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. Nucleation is based on Wexler et al. (1994). The CBM-Z-MOSAIC 8 bin scheme is not coupled to a ~~secondary organic aerosol (SOA) scheme in our version of WRF-Chem (3.5.1).~~ ~~Furthermore, According to Bessagnet et al. (2008), 75–95 % of annually averaged SOA in Europe is associated with biogenic sources. However, biogenic VOC (Volatile Organic Compounds) emissions are relatively low in Europe during the months of March and April (Karl et al., 2009). In addition, Bessagnet et al. (2008) point out that SOA concentrations are much lower in northern Europe than in other European regions. For all of these reasons, and since current SOA mechanisms are still highly uncertain (e.g. Hodzic et al., 2010; Gustafson et al., 2011), and SOA formation is likely to be low in Europe and at high latitudes in early April. Because of this, our simulation did not include SOA formation~~ the present simulations do not include SOA formation. However, we note that Frossard et al. (2011) determined that SOA formation

contributed in part to the organic aerosol fraction in the Scandinavian marine boundary layer during the period of the POLARCAT-France flights (April 2008), and that our simulations cannot reproduce this contribution. MOSAIC considers aerosols as internally mixed in each bin, and in our simulations optical properties are calculated using volume averaging.

The simulation domain, focused on the POLARCAT-France spring flights, is shown in Fig. 1 and covers Europe north of 40° N and west of 70° E. The spatial resolution is $30\text{ km} \times 30\text{ km}$ horizontally, with 50 vertical levels up to 50 hPa. Anthropogenic emissions were taken from the HTAPv2 $0.1^{\circ} \times 0.1^{\circ}$ inventory (http://edgar.jrc.ec.europa.eu/htap_v2/index.php?SECURE=123). HTAP VOCs are given as a bulk VOC mass, and are distributed into CBM-Z emission categories assuming the speciation of UK VOCs determined by Murrels et al. (2010). Time profiles are applied to anthropogenic emissions to account for the daily and weekly cycle of each emission sector (Denier van der Gon et al., 2011). Fire emissions are from the FINN v1 inventory (Wiedinmyer et al., 2006, 2011), and are injected in altitude by an online plume rise model described in Freitas et al. (2007). Figure 2 shows black carbon (BC), organic carbon (OC), and sulfur oxides (SO_x) emissions during our simulation, from both anthropogenic sources (panels A, B and C) and biomass burning sources (panels D, E and F). In-domain biomass burning emission totals are 13 kilotons (kt) for SO_x , 12 kt for BC and 75 kt for OC. For anthropogenic emissions, **emission totals in-domain emission totals from HTAPv2** are 575 kt for SO_x , 21 kt for BC and 46 kt for OC. Anthropogenic emissions are stronger in Western and Central Europe, especially in Poland and Slovakia. Biomass burning emissions are located in the eastern part of the domain, because of intense agricultural fires in Ukraine, Russia and Kazakhstan during early April 2008 (Warneke et al., 2009). Biogenic emissions are calculated online in WRF-Chem by the model MEGAN (Guenther et al., 2006). Finally, sea salt aerosol emissions are calculated online, while mineral dust emissions are not included.

Boundary and initial meteorological conditions in the simulation are given by the global NCEP Final Analysis (FNL), and WRF-Chem temperature, humidity and winds are nudged every 6 hours to the reanalysis above the atmospheric boundary layer. Trace gases and

aerosol initial and boundary conditions (updated every 6 h) are taken from the global chemical transport model [MOZART-4](#) [MOZART-4](#) (Emmons et al., 2010).

WRF-Chem simulations include a control run (CTL) from 00:00 UTC 1 April to 00:00 UTC 12 April using the model and emissions as described above. We also perform 4 sensitivity simulations for the same period to investigate the sources, processes along transport and regional impacts of aerosols sampled during POLARCAT: (1) removing the HTAPv2 emissions (NOANTHRO), (2) without biomass burning emissions (NOFIRES), (3) a simulation with wet scavenging turned off (NOWETSCAV), and (4) [a simulation with the aerosol direct interaction with short wave radiation turned off disabled, thus switching off the direct and semi-direct aerosol effects](#) (NODIRECT). The NOANTHRO and NOFIRE simulations are used [in Sect. 5.1](#) to estimate the contribution of European anthropogenic and biomass burning emissions to Arctic aerosols measured during POLARCAT. The NOWETSCAV simulation allows us to quantify [in Sect. 5.2](#) the magnitude of the wet scavenging of aerosols during their transport from Europe to the Arctic. The NODIRECT simulation is used [in Sect. 6](#) to estimate the direct and semi-direct shortwave radiative effect (DSRE) of aerosols associated with this transport event.

To compare simulations with airborne LIDAR measurements, modeled backscatter ratio profiles at the plane position are calculated by using the aerosol backscattering coefficient at 400 nm simulated by WRF-Chem. This coefficient is computed from the method of Toon and Ackerman (1981), using a bulk, volume averaged, refractive index derived from the modeled size distribution (Bond et al., 2006). The backscattering coefficient is then estimated at 532 nm by using the simulated Angström exponent, and the effect of aerosol transmission is ignored because aerosol optical depths of observed layers were low ($< 4\%$) during POLARCAT-France (Adam de Villiers et al., 2010). The backscatter ratio is calculated following the definition in Sect. 2.1, where the molecular backscattering is estimated by an empirical formulation of the Rayleigh scattering (Nicolet, 1984) using meteorological profiles from the CTL simulation.

2.3.2 FLEXPART-WRF

We also use FLEXPART-WRF, a Lagrangian particle dispersion model (Brioude et al., 2013) adapted from the model FLEXPART (Stohl et al., 2005), to study air mass origins and transport processes using WRF meteorological forecasts. In this study, we use FLEXPART-WRF in backward mode to study the origin and transport pathways of plumes measured during the POLARCAT-France spring campaign, and to provide insight into the WRF-Chem representation of aerosols. The meteorological fields from the WRF-Chem simulation CTL described in 3.1 are used as input. Every minute, 10 000 particles are released along the aircraft flight tracks in a volume $10 \text{ km} \times 10 \text{ km}$ (horizontally) and 400 m (vertically). Each of the simulations is run backwards for 7 days to track the air mass origin over the source regions of interest (transport times are typically less than 7 day). Specifically, we use FLEXPART-WRF Potential Emission Sensitivity (PES) to study source–receptor relationships for air measured by the ATR-42 as part of the POLARCAT-France spring flights.

3 Meteorological context during the spring POLARCAT-France campaign

Long-range transport of aerosol from Europe to the Arctic is usually associated with specific synoptic meteorological situations over Europe, causing large scale meridional transport (e.g. Iversen and Joranger, 1985). In order to investigate the origin and transport of aerosols measured during the POLARCAT-France spring campaign, the synoptic meteorological situation during the campaign as represented by WRF-Chem is shown in Fig. 3. Specifically, WRF-Chem simulated geopotential height contours and wind arrows (700 hPa) are shown from 6 to 11 April 2008. A similar figure showing wind speed at 700 hPa instead of geopotential height is shown in the Supplement, Fig. S1. A low pressure over the North Sea and a high pressure over southwestern Russia and Kazakhstan caused southerly winds over Central and Eastern Europe from 6–8 April. On 8 April, the North Sea low pressure moved over the Baltic Sea, pushing those southerly winds deeper into the Scandinavian Arctic. On 9 April, the low pressure weakened and moved over Finland, while a deep trough formed

over the Kara Sea, stopping northward transport and producing strong westerly winds over Europe and western Russia through the end of the aircraft campaign on 11 April.

Aerosols and other pollution are transported from lower latitudes in Europe in these synoptic meteorological systems, which determine the main pollution transport pathways. We show vertically integrated black carbon as a proxy for pollution transported during this time period in Fig. 4 (CTL simulation). The intersection of the low over the North Sea and the high located over Russia lead to the northward transport of a large polluted air mass from Central and Eastern Europe. A portion of this air mass was carried eastward at mid-latitudes, while another portion reached Arctic Scandinavia on 8 to 9 April. This polluted air mass was sampled by POLARCAT-France flights on 9, 10 and 11 April 2008, the flights that are the main focus of this study. However, this air mass did not penetrate deep into the Arctic and mix significantly with Arctic air due to the position of the polar front (Ancellet et al., 2014). On 10–11 April, the Arctic outflow intensified in the Barents and Norwegian Sea, slowly transporting the polluted European air back to lower latitudes. On 10–11 April, pollution (represented as elevated BC) can be seen entering the simulation domain from the northern boundary over Svalbard (in our simulations via the [MOZART-MOZART-4](#) boundary conditions), and crossing the POLARCAT flight track on 11 April. This last polluted air mass is not the focus of the present study and has been identified [by](#) as a mixed anthropogenic and biomass burning plume originating from northeast Asia. It has already been studied in detail by Adam de Villiers et al. (2010) and Quennehen et al. (2012).

4 Model validation

Results from WRF-Chem are compared to POLARCAT-France [1-s resolution](#) measurements of temperature, relative humidity, wind speed, and wind direction (CTL simulation) for the POLARCAT-France flights included in our study. This comparison is presented in Fig. 5. Modeled and measured quantities are in good agreement with the exception of fine scale features that are not reproduced by the model due to the horizontal grid spacing (30 km). In particular, we note that relative humidity (RH) is well reproduced by the

model ($R^2 > 0.88$). Pilinis et al. (1995) showed that RH, through aerosol water uptake, is a key parameter for modeling aerosol optical properties. The main discrepancies are between the measured and modeled wind speeds on 10 April 2008, in-which-when high winds were observed below 1 km (middle portion of the flight) over the Norwegian Sea. However, discrepancies between modeled and measured wind speeds in the marine boundary layer over the Norwegian Sea during this portion of the flight-flight do not impact the results for the pollution events we focus on, which were encountered higher up in the Scandinavian free troposphere and were emitted over continental Europe. The model performance in the Arctic troposphere indicates that the model captures the changing meteorological conditions in the European Arctic at the end of the POLARCAT-France spring campaign (discussed earlier in Sect. 3). This provides confidence that plume transport and dispersion are adequately represented to study aerosol transport and processing.

We evaluate model performance over the European source regions by comparing background aerosol levels from the EMEP network with model results (CTL simulation) extracted at the station locations. Figure 6 shows the comparison for $\text{PM}_{2.5}$, SO_4^- , NO_3^- , and NH_4^+ , daily averaged for all stations. Error bars show the standard deviation between stations for both measured and modeled aerosols. Overprediction of aerosols on 1 April for $\text{PM}_{2.5}$, NO_3^- , and NH_4^+ correspond to positive biases for these species in the initial conditions (MOZART4MOZART-4), but WRF-Chem results are in better agreement with measurements after one day of simulation. This first day is considered as model spin-up, and is excluded from further analysis. We evaluate the model performance in reproducing European background aerosol levels in terms of Normalized Mean Bias (NMB). It is defined as
$$\text{NMB} = 100\% \times 1/N \times \sum_{i=1}^N (M_i - O_i) / O_i$$
 where M_i and O_i are modeled and observed daily values, averaged over all sites, and the summation is over the $N = 10$ days between 2 and 11 April. $\text{PM}_{2.5}$ levels are well reproduced by the model (NMB = -0.9%). There are more significant differences in measured and modeled aerosol composition: while SO_4^- agrees well with measurements (NMB = -0.6%), NO_3^- (NMB = $+107\%$) and NH_4^+ (NMB = $+53\%$) are overestimated. This suggests that the overestimation of NO_3^- and NH_4^+ might be com-

pensated [in terms of overall mass](#) by an underestimation of organic carbon (OC) aerosols, [resulting in relatively good PM_{2.5} agreement](#). Due to a lack of available OC measurement from EMEP stations for this period, this hypothesis cannot be verified. If we use the very limited EMEP OC data (5 stations, 67% coverage), we find that OC is indeed underestimated for those stations (NMB = -38%). This underestimation could be caused, in part, by the fact that SOA is not included in our model run. [Since SOAs can be formed by the oxidation of VOCs by gas phase NO₃, it is also possible that the lack of SOA is related to the overestimation of nitrate aerosols in our simulations](#). However, we [also](#) note that previous studies including SOA can report errors on OC of the same magnitude or larger (e.g. -74% in Tuccella et al., 2012, [who attribute this deficiency in modeling OC to an incomplete description of SOA formation in their mechanism](#)).

The overestimation of NO₃⁻ and NH₄⁺ and underestimation of OC by WRF-Chem in Europe were also seen in the simulations of Tuccella et al. (2012), using different emissions as well as gas and aerosol schemes. That study suggested the discrepancy was due to missing aqueous reactions causing an underestimation of sulfate formation, leading to less neutralization of ammonium by sulfate and favoring the formation of ammonium nitrate (see Meng et al., 1997). It also highlighted the possible role of uncertainties in the simplified wet scavenging scheme used for that study. Our study includes a more complete wet scavenging scheme and the full range of aqueous reactions included in MOSAIC, keeping in mind that cloud/aerosol interaction processes in MOSAIC are only accounted for in dynamically resolved clouds, which should be underestimated in our simulation (30 km horizontal resolution). The inclusion of these processes, and the use of different anthropogenic emissions (EMEP in Tuccella et al. (2012) vs. HTAPv2 in the present study) can explain the better agreement on sulfate compared to Tuccella et al. (2012). However, this better agreement also means that, in our case, sulfate concentrations do not drive the overestimation of modeled ammonium and nitrate. Using EMEP measurements of ammonia (19 stations) and NO_x (10 stations), we found that NH₃ is overestimated by a factor of 2 in our simulation (NMB = +108%) while NO_x is slightly underestimated (NMB = -23%). This overestimation

of NH_3 could cause ~~of~~ an enhanced formation of ammonium nitrate, which would explain the model overestimation of ammonium and nitrate.

While the CTL simulation is able to reproduce $\text{PM}_{2.5}$ levels observed in source regions, this good performance is due in part to compensating effects between different chemical components of the aerosols. The hygroscopy-bulk hygroscopicity of OC ($\kappa = 0.14$) is lower than the one for NO_3^- and NH_4^+ ($\kappa = 0.5$) in MOSAIC. This means that the underestimation of OC in our simulation might lead to overestimated aerosol activation in clouds and wet scavenging. However, refractive indices for OC, NH_4NO_3 and $2(\text{NH}_4)_2\text{SO}_4$ are close (1.45, 1.50, 1.47 in MOSAIC), meaning that compensation between these different components should not have a strong impact on modeled aerosol optical properties, and that our model represents European aerosols sufficiently well to investigate their the direct and semi-direct aerosol radiative effects in the Arctic.

5 The origin and properties of springtime aerosols during POLARCAT-France

In this section, modeled aerosols in the Arctic are compared with POLARCAT-spring measurements, to investigate in detail the aerosol transport event from Europe to the Arctic. We combine WRF-Chem simulations with FLEXPART-WRF to identify the source regions and transport pathways of plumes sampled during the campaign, and show how they impact processes along transport and the vertical structure of Arctic pollution. First, aerosol particles detected in plumes in April 2008 are described in terms of mass concentrations, chemical composition and number size distributions. The role of transport pathways and wet scavenging along transport on those properties is also investigated. Aerosol optical properties are then used to quantify the vertical distribution of aerosols as a function of their emission sources.

5.1 Modeling aerosols measured in situ on 9, 10 and 11 April 2008

POLARCAT-France measured (in-situ) $\text{PM}_{2.5}$ is compared with modeled $\text{PM}_{2.5}$ interpolated in space (model results using hourly output) along the flight tracks on 9, 10, and

11 April 2008 (Fig. 7). The time series of measured $\text{PM}_{2.5}$ shows plumes containing enhanced aerosols were encountered during the flights. Aerosol mass in plumes ranged from 3 to $16 \mu\text{g m}^{-3}$, while background-baseline levels were $\sim 1 \mu\text{g m}^{-3}$. It should be noted that clean-Arctic-background-unpolluted air and marine boundary layer air were less frequently sampled due to the planned flight patterns, which targeted anthropogenic and biomass burning influenced plumes. Gray shading denotes periods when in-situ measurements are not available, usually due to the presence of clouds.

Air mass origins indicated on Fig. 7 are determined using a combination of WRF-Chem and FLEXPART-WRF (simulations described below). The influence of anthropogenic and biomass burning emissions on the flight track is estimated using the NOANTHRO and NOFIRE sensitivity runs. Specifically, this influence is deemed significant if aerosol mass increased by more than 20 % upon including either anthropogenic or biomass burning emissions, according to the ratios $[\text{CTL PM}_{2.5}]/[\text{NOANTHRO PM}_{2.5}]$ and $[\text{CTL PM}_{2.5}]/[\text{NOFIRE PM}_{2.5}]$. The values of these ratios along the three flight tracks are presented in the Supplement, Fig. S2. We used a threshold of 20 % to highlight the difference between air masses significantly influenced by biomass burning (BB) and air masses mostly influenced by anthropogenic emissions. This threshold excludes air masses weakly influenced (5 to 15 %) by BB on 10 and 11 April (as seen on Fig. S2) and identifies air masses significant influenced by BB, up to 30–40 %. We used the same threshold of 20 % for anthropogenic plumes for consistency. On Fig. 7, pink shading indicates that the modeled $\text{PM}_{2.5}$ are influenced by European Anthropogenic emissions. Yellow shading indicates portions of the flight influenced by both biomass burning and anthropogenic emissions (mixed plumes). It should be noted that portions of the flight track that are influenced by biomass burning emissions are also influenced by anthropogenic emissions. Green shading indicates that the modeled air mass is significantly influenced by the domain northern boundary conditions (i.e. air transported from Asia). This influence is identified using FLEXPART-WRF, run in backwards mode with particles released every minute along the flight tracks ($30 \text{ km} \times 30 \text{ km}$ horizontally by 400 m vertically). When the FLEXPART-WRF retroplume mean trajectory passes closer than 5 grid cells (150 km) from the northern end of the domain, the air mass is considered

as influenced by the northern boundary conditions. The typical transport pathway of such a plume is shown in the Supplement, Fig. S3. Finally, white shading indicates air masses that are not attributed to a specific source using the methods described above and are referred to as **background-unpolluted** air.

In the free troposphere, the model is able to reproduce the **background-baseline** $\text{PM}_{2.5}$ levels and the main peaks observed in European air masses for all three flights. The Normalized Mean Bias for $\text{PM}_{2.5}$ for all three flights, excluding **background-Arctic-unpolluted** air and boundary condition air, is +8.8%. Peaks attributed to European anthropogenic emissions are reproduced, although the model cannot capture some small-scale features due to its resolution. At the end of the 9 April flight, two concentrated plumes were sampled in situ around 12:00 and 12:15 UTC. The model identifies these plumes as mixed (anthropogenic/biomass burning), meaning that significant ($> 40\%$) enhancements in modeled $\text{PM}_{2.5}$ at these times are due to biomass burning or anthropogenic European emissions. The first $\text{PM}_{2.5}$ peak is underestimated by the model (around 12:00 UTC), and the second plume (around 12:15 UTC) is located 1.5 km too low in altitude. This may be due to uncertainties in the injection height for fires or in the intensity and timing of the emissions. However, the issue does not appear to be systematic in our simulation because mixed plume peaks and enhancements are correctly represented during the 11 April flight. Modeled anthropogenic $\text{PM}_{2.5}$ are underestimated below 1 km at the beginning and end of the 11 April flight above Sweden (discussed in detail in Sect. 5.3). Plumes coming from the northern domain boundary, which are not studied in detail here, reflect **the aerosols present in the MOZART-4—a general underestimation of aerosols in the MOZART-4** simulation used as the boundary conditions **and point to a general underestimation, a feature seen in many global aerosol transport models**. On 9 April, WRF-Chem also reproduces a large $\text{PM}_{2.5}$ peak located in the marine boundary layer. This peak is composed of more than 95% sea salt in the model, and corresponds to sea spray uplifted by the strong 20 m s^{-1} winds present in the marine boundary layer in the region of the flight.

The modeled composition of $\text{PM}_{2.5}$ aerosols in anthropogenic and mixed polluted air masses is presented in Table 2. On 9 and 10 April, anthropogenic plumes are mostly

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composed of nitrate, sulfate and ammonium aerosol. Mixed plumes contain relatively less nitrate, but more sulfate, organic carbon, and black carbon. The proportion of sulfate is higher in mixed plumes than in anthropogenic plumes, despite the fact that sulfate and SO₂ emissions from biomass burning emissions are low. We show in the next section focused on plume origins that the proportion of sulfate is high for mixed plumes because they originate in a region of high anthropogenic SO₂ emissions. On 11 April, the composition of anthropogenic plumes and mixed plumes are similar, except for organic carbon, which is still lower in anthropogenic plumes. In Sect. 4, we showed that the model was overestimating nitrate and ammonium at the surface, while probably underestimating organic matter in the European source regions. Measurements of aerosol chemical composition are not available along the POLARCAT-France flights, but ~~we can assume that similar biases apply to the modeled aerosol composition in the Arctic. were determined during other POLARCAT campaigns in other parts of the Arctic. In-situ measurements during other campaigns generally indicate less nitrate and more organic matter (OM) in Arctic aerosols. For example, Brock et al. (2011) found 78 % OM and 20 % NO₃⁻ in biomass burning aerosols in the Alaskan Arctic during ARCPAC (32 % and 1 % for anthropogenic plumes). Airborne AMS measurements in the summer in Greenland during POLARCAT-France (Schmale et al., 2011) also indicate very low NO₃⁻ concentrations (below the detection limit) and high proportions of OM (50 to 90 %) in polluted 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 % OM, 60 % SO₄⁻ and 1 % NO₃⁻ in aerosols found in the Scandinavian marine boundary layer. This comparison with other POLARCAT data also indicates that in our simulations, nitrate aerosols might have been formed at the expense of organic matter, probably due to the lack of a SOA mechanism.~~ The proportion of black carbon modeled in the present study is 2.5 % in anthropogenic air masses (2.6 % for submicron particles), and 3 in-mixed plumes (3.1 % for submicron particles). These values are comparable with results from Brock the study of Brock et al. (2011), a study that which found on average 2.4 % submicron mass of BC in anthropogenic plumes and 3.5 % in fire plumes in the Alaskan Arctic during spring 2008.

~~Aerosol optical and microphysical properties are very sensitive to their~~ We evaluate model predictions of aerosol size distributions, which are known to be important for the optical properties (e.g. Boucher, 1998) presented in sections 5.3 and 6. It is also important to note that activation in clouds, which is outside the scope of the present study, is also sensitive to aerosol size distributions (~~Boucher, 1998;~~ Dusek et al., 2006). ~~To ensure that aerosol impacts are treated accurately in the CTL simulation, modeled aerosol number~~ 1996). Plumes for which we compare modeled and measured size distributions are validated against in-situ measurements for selected plumes. Those plumes are indicated in each panel on ~~indicated by ticks in~~ Fig. 7 ~~by ticks~~ (referring to the modeled aerosol peak). 4 anthropogenic plumes (I, J, M, N) and 3 mixed plumes (K, L, O) are investigated. In the case of plume K, the modeled plume peak is located 1 km lower in the model than in observations, which results in it being displaced later in time along the flight track. For this plume, we compare the modeled and measured plumes using the peak aerosol mass encountered in the model (12:19 UTC) and measurements (12:14 UTC) respectively. This comparison is shown in Fig. 8. It indicates that the model adequately represents the aerosol size distributions with three exceptions. First, the model overestimates the number of ~~large particles~~ particles larger than 200 nm in the 9 April anthropogenic plumes (I, J). Second, the model cannot be compared to measurements in the smallest MOSAIC bin (aerosols 39 to 78 nm), due to the fact that the model does not resolve explicitly nucleation, but relies on a parameterization for nucleation and growth of particles with diameters less than 39 nm. Third, number concentrations are overestimated in the 2nd smallest MOSAIC bin (aerosols 78 to 156 nm) for mixed plumes (K and L–O) but not for anthropogenic plumes. We show in Sect. 5.2 that mixed plumes are ~ 2 days older than anthropogenic plumes. This means that this overestimation is probably caused by underestimated growth processes, which have the largest impact on older plumes. However, aerosol optical properties are mostly sensitive to particles in the accumulation mode, which is correctly reproduced for all plumes (Stokes diameter ranges for these modes are 90–500 nm for the anthropogenic plumes, and 110–700 nm for the fire plumes, Quennehen et al., 2012).

5.2 Origins and transport pathways of anthropogenic and biomass burning plumes sampled during POLARCAT-France

Different types of aerosols transported to the Arctic during POLARCAT-France display different physical properties and vertical distributions. We investigate how different plume origins and transport pathways result in different aerosol properties in the Arctic. We focus on the role of wet scavenging during transport, which is the largest source of uncertainty in the representation of Arctic aerosols (Schwarz et al., 2010; Browse et al., 2012). Figure 9 shows typical plume transport pathways of an anthropogenic plume (plume J, Fig. 9a and c) and a mixed plume (plume K, Fig. 9b and d) measured during the campaign. Figure 9a and b shows the 0–20 km column of FLEXPART-WRF PES integrated for 7 days for both plumes. It indicates that anthropogenic plumes were mostly influenced by sources in central Europe 2–3 days prior to the measurements, while the mixed plume is 3 to 5 days old and under the influence of emissions in a large **large**-region over eastern Europe and western Russia. This region corresponds to the location of agricultural fires in early April 2008, as well as significant anthropogenic emissions, especially of SO₂, as seen in Fig. 2. The larger age of mixed plumes explain why their size distribution is shifted toward larger sizes than younger anthropogenic plumes, as discussed in Quennehen et al. (2012).

Figure 9c and d show the mean altitude for each plume as a function of age. The anthropogenic plume experienced a rapid uplift from 1.5 to 6.5 km over Poland and the North Sea on 7 or 8 April, associated with the surface low over this region, while the mixed plume was transported to the Arctic below 2 km and slowly uplifted. Between 9 April and 11 April, FLEXPART-WRF trajectories (not shown here) inform us that mixed plume K mixed with air from fresher anthropogenic plumes I and J. This mixing explains why the chemical composition of the 11 April mixed plumes, showed in Table 2 and discussed above, is intermediate between 9 April mixed plume K and the 9 April anthropogenic plumes I and J.

The magnitude of wet scavenging along transport, also represented **on-in** Fig. 9c and d, is estimated using the difference between CTL PM₁₀ minus NOWETSCAV PM₁₀ along the retroplumes positions. As expected, strong PM₁₀ depletions, reaching **-210** μg m⁻³

(−74 %) are associated with precipitation during uplift of the anthropogenic plume in the frontal system over Poland, between 37 and 46 h before it was measured. Although the mixed plume does not experience such a rapid uplift, aerosols are also scavenged by rainout over Finland, between 35 and 45 h before sampling, decreasing PM_{10} levels by $17 \mu\text{g m}^{-3}$ (−55 %). The accumulated precipitation in the simulation, compared to the E-OBS European daily gridded precipitation dataset (Haylock et al., 2008), shows that while WRF-Chem correctly reproduces the precipitation patterns observed during this period, it generally underestimates their intensity (see Supplement, Fig. S4). However, we have shown that average $\text{PM}_{2.5}$ levels are well reproduced in the source regions and in the Arctic, indicating that losses along transport are relatively well reproduced. This could be explained by compensations between underestimated precipitations and an overestimated wet scavenging rate in our simulation. An overestimation of the wet scavenging rate could be caused by the overestimated hygroscopy of the modeled aerosol, which contains too much ammonium and nitrate, and not enough organic matter.

5.3 Vertical aerosol distributions: 9 April 2008

The vertical structure of aerosol layers transported to the Arctic is often complex (Brock et al., 2011), and the vertical distribution of absorbing aerosol layers can have a large influence on their radiative effects (e.g. Meloni et al., 2005; Raut and Chazette, 2008). Here, the modeled vertical structure of aerosol layers in the Arctic troposphere is evaluated using the pseudo backscatter ratio at 532 nm (PBR) measured by the airborne LIDAR shooting at nadir. The measured PBR is represented in Fig. 10b for the 9 April flight, clouds and data below clouds are masked in white. The altitude of the aircraft, which was going north to south and returning to Kiruna, is shown as a black line on panels B to E. We choose to show the 9 April flight because modeled low-level pollution is not influenced by the model northern boundary conditions on this day. The model to observations comparison is therefore not affected by the performance of the global model [MOZART4](#)[MOZART-4](#). Figure 10a shows the $\text{PM}_{2.5}$ measured in situ by the aircraft during the same period. The $\text{PM}_{2.5}$ and LIDAR-derived PBR just below the aircraft present a very similar evolution: the $\text{PM}_{2.5}$ and

PBR signals are enhanced during the whole leg between 4 and 5 km, at the aircraft altitude and just below. This good correlation ($r^2 = 0.86$, see Fig. S5) between aerosol mass and optical properties allows us to validate aerosol concentrations vertical distributions through their optical properties.

The PBR at 532 nm is compared to cross-sections of the simulated backscatter ratio (Fig. 10c), simulated $\text{PM}_{2.5}$ (Fig. 10d) and simulated aerosol number concentration (Fig. 10e) extracted along flight tracks from the WRF-Chem simulation. The magnitude of the PBR is correctly reproduced, with background regions between 1 and 1.1, and visible aerosol layers reaching values of 1.3 to 1.5. Peak intensities in plumes transported to the Arctic region tend to be underestimated by the model, as the modeled plumes are too diluted vertically. Plume locations are reasonably well reproduced with an enhanced layer at 5 km during the whole flight leg, and two main layers at lower latitudes and altitudes, between 1.5–2 and 3–4 km. One enhanced layer measured between 11:30 and 11:50 UTC at 1 km is missing from the modeled **ASPR-PBR** cross-section because it is displaced ~ 50 km to the southwest in the simulation (see Supplement, Fig. S5S6). This displacement is probably due to the cumulative effect of small errors on wind speed and wind direction over the 3 to 5 days of long-range transport. The model underestimates the PBR in the intense layer measured in situ and by the LIDAR at 5 km at 12:00 UTC, which is in agreement with the underestimation observed on $\text{PM}_{2.5}$ levels previously described in Fig. 7. This layer, identified as a 5-day-old mixed plume in the model, features low $\text{PM}_{2.5}$ but high aerosol number concentration (Fig. 10e), suggesting it is mostly composed of small particles. This means that the discrepancy in this layer probably corresponds to underestimated **condensation processes in the aerosol model** 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 investigate the vertical distribution of modeled anthropogenic and biomass burning aerosols during this profile, and the impact of wet scavenging on the vertical distribution.

Figure 11 shows the sensitivity of the $\text{PM}_{2.5}$ vertical cross section to anthropogenic emissions (Fig. 11a), biomass burning emissions (Fig. 11b) and wet scavenging (Fig. 11c). During the 9 April flight, anthropogenic emissions have the largest influence in the mid to upper troposphere, above 4 km and in the PBL and lower troposphere, below 2 km, while the impacts of biomass burning emissions are more pronounced between 2 and 4 km. Figure 11b confirms that the plume missing at 5 km in Fig. 10c is indeed due to biomass burning emissions, but the associated enhancement above background is very low, around $1 \mu\text{g m}^{-3}$. According to Fig. 11c, this low enhancement is not due to high wet scavenging in this layer. As discussed before, this confirms that the underestimation of $\text{PM}_{2.5}$ in this layer may be due to insufficient [condensation growth by condensation in this plume](#). The impact of wet scavenging is the strongest for the lower level mixed pollution, as discussed in the case of plume K in Fig. 9d. It is negligible in biomass burning layers located between 2 and 4 km, and strong relatively to total $\text{PM}_{2.5}$ in the southernmost and low-altitude anthropogenic layer.

6 Impacts of European aerosol transport on the Arctic

Results presented so far give us confidence in the way this transport event is represented in our simulation in terms of meteorology, $\text{PM}_{2.5}$ levels, size distributions, spatial extent and vertical structure of the plumes. We now investigate the regional impacts of this transport event in the European Arctic region. Figure 12 shows the average vertical profiles of the modeled anthropogenic and biomass burning contributions to $\text{PM}_{2.5}$ (total and chemically speciated) north of the Arctic circle (within the model domain) during the period from 00:00 UTC 8 April, to 00:00 UTC 12 April. The very low aerosol concentrations are due to area-weighted averaging of European enhancements confined in the lower Scandinavian Arctic with the rest of the clean Arctic region contained in the domain. Because of this, we will not discuss the absolute enhancements and instead focus on relative values. This average profile shows the same general features [than what was as were](#) observed in-situ and by LIDAR during POLARCAT-France, with anthropogenic emissions separated between

a low altitude (1.5 km) and a high altitude (4.5 km) contribution, and biomass burning emissions impacting intermediate altitudes (2.5–3 km). Different species display different vertical structures: for the anthropogenic contribution, BC, OC, and SO_4^- are enhanced at low altitudes. This corresponds to the mixed layers from eastern Europe and Russia. High altitude anthropogenic plumes from central Europe contain enhanced NH_4^+ , NO_3^- and BC. Biomass burning plumes contain larger mass fractions of BC and OC than anthropogenic plumes, and BC and OC influence lower altitudes than other $\text{PM}_{2.5}$ species from biomass burning. These results are in agreement with earlier studies by Stohl et al. (2007) and Lund Myhre et al. (2007), who analyzed cases of transport of biomass burning plumes from eastern Europe to the Arctic in spring 2006. Using FLEXPART simulations and LIDAR measurements, they showed that biomass burning aerosols were mostly confined below 3 km altitudes in the Arctic. Fischer et al. (2011) investigated aerosol transport from the mid-latitudes to the Arctic during April 2008 with the global chemical transport model GEOS-Chem, and found that in the high Arctic (75–85° N), NH_4^+ and SO_4^- were sensitive to European anthropogenic emissions at all altitudes, with a peak sensitivity between 2 and 5 km.

Pueschel and Kinne (1995) have shown that layers of aerosols containing black carbon, even with very high single scattering albedos (0.98), could warm the atmosphere over snow or ice covered surfaces. 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 ~~should~~may contribute to enhanced local atmospheric heating rates in this region. ~~Figure 13a shows~~ (Flanner, 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 flux calculated online by the Goddard shortwave module within WRF-Chem, in the CTL simulation minus the NODIRECT simulation. Because WRF-Chem upward radiative fluxes are by convention always negative, positive DSRE values at TOA indicate heating of the surface–atmosphere column. The DSRE is averaged over the period

from 00:00 UTC 8 April to 00:00 UTC 12 April. In-domain anthropogenic and biomass burning emissions are considered significant if the $\text{PM}_{2.5}$ column sensitivity to anthropogenic and biomass burning emissions (shown in Fig. 13b) exceeds 50 % of the total column of CTL $\text{PM}_{2.5}$. We added this condition to exclude from our calculation of the DSRE the areas where the dominant contribution is due to aerosols originating from the boundary conditions (i.e. the Asian plume), from natural emissions (i.e. sea salt) or from background levels.

As expected, the DSRE is negative over land and ocean where snow and ice cover are low, but positive over regions with high snow and ice covers (see the snow and ice cover map on Fig. 13c). The 4 day average value of the DSRE at TOA north of 60°N in regions significantly influenced by European pollution is shown in Table 3. In addition to the total average effect north of 60°N , we compute values for the DSRE over surfaces with extensive snow and ice cover ($> 90\%$), and over the ocean surface. On average, the European aerosols have a cooling effect north of 60°N (-0.98 W m^{-2}). Over snow and ice, the average DSRE is $+0.58\text{ W m}^{-2}$, peaking near $+2\text{ W m}^{-2}$ over a large region in northern Scandinavia where aerosol optical depths (AOD) are the highest (~ 0.5 at 400 nm). The DSRE is much lower over the Russian snowpack east of 42°E because the European mixed air mass in this region is either optically shallow (AOD from 0.05 to 0.2) or is located below clouds. Over the Arctic seas, the DSRE is negative due to the lower albedo of the ocean surface. The calculated DSRE in oceanic regions north of 60°N influenced by the European plumes is -1.5 W m^{-2} . Minimum values reach close to -5 W m^{-2} over the Norwegian Sea close to the coast of Norway, where the cloud cover is the lowest, as shown in Fig. 13d.

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 in different locations within the Arctic. We summarize the other studies for comparison, but we leave it to future studies to draw broader conclusions about whether these results are representative of wider spatial and temporal scales. Brock et al. (2011) calculated a direct radiative effect of $+3.3\text{ W m}^{-2}$ over snow at TOA for the average of 10 typical polluted profiles measured during the ARC-PAC campaign, not taking the semi-direct effect into account. Maximum modeled BC in WRF-Chem along the POLARCAT-France flight tracks is 150 ng m^{-3} (anthropogenic) and

260 ng m⁻³ (mixed fire/anthropogenic), which are comparable with the average BC values reported for anthropogenic (148 ng m⁻³) and fire plumes (312 ng m⁻³) in Brock et al. (2011). This means that on average, the BC values for pollution-influenced plumes in our simulation are lower than values reported by Brock et al. (2011). Quinn et al. (2007) found a similar direct radiative effect value of +2.5 W m⁻² over snow at TOA for the average polluted conditions encountered during the Arctic haze maximum at Barrow. Those results were obtained at solar noon, in clear sky conditions, over snow and in polluted regions only, conditions that lead to a maximum direct effect. Using a similar approach, we compute the DSRE in regions influenced by European pollution, close to noon (11:00 UTC), and above high snow covers (> 90%). This results in an average DSRE of +1.9 W m⁻² north of 60° N. If we exclude the snowpack in Russia, east of 42° E, the average DSRE reaches +3.3 W m⁻². These values are in agreement with results from Brock et al. (2011) and Quinn et al. (2007). It should be noted that our retrievals are done in all-sky conditions and not exactly at local solar noon, introducing a slight low bias. Including the semi-direct effect in our calculations might have introduced a warming bias, which would be limited by the nudging of WRF-Chem temperature, relative humidity and wind speed towards FNL reanalyzes in the free troposphere. We verified that differences in cloud cover between the NODIRECT and CTL simulations were limited in magnitude and extent, with only a few local points over the sea affected (below 10% cloud cover change for the 8 to 12 April average), that mostly cancel each other out when regionally averaged.

Lund Myhre et al. (2007) calculated the direct forcing of biomass burning aerosols transported from Europe to the Arctic in late April and early May 2006 from space borne aerosol optical depth measurements. For those exceptionally intense plumes, they found that the cooling direct effect at TOA reached -35 W m⁻² over the regions with the highest AOD in the Barents Sea, while the maximum warming direct effect over snow was limited to +5 W m⁻² over Svalbard. Keeping in mind that our results are not directly comparable because of the different times of year and different averaging periods, we found a 4 day average direct and semi-direct effect reaching maximum values of +2 W m⁻² over snow-covered Scandinavia, and maximum cooling values of -5 W m⁻² over the Norwegian Sea.

Several reasons could explain this different balance between warming and cooling effects. In our case, modeled European plumes contained higher levels of black carbon (2.5 to 3% of submicron aerosol mass) than the measured value used in the study of Lund Myhre et al. (1.98%). The transport event studied here also featured a high altitude anthropogenic plume that would have a local warming effect above the high albedo low-level clouds. The inclusion of the semi-direct effect in our study might have also played a limited role.

At the surface, the direct aerosol effect causes local cooling for all types of land surfaces, ~~but we also showed 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 Fig. 12 that BC was ~~especially~~ enhanced at the surface in anthropogenic ~~and biomass burning~~ plumes, which could lead to surface warming through ~~snow-albedo~~ the effects of BC deposited on snow. ~~This effect is not taken into account in our simulation, but an earlier study by Wang~~ 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 during in spring 2008 (April–May), significant levels of anthropogenic BC (1 to $5 \text{ mg C m}^{-2} \text{ month}^{-1}$) were deposited on snow in Northern Europe, leading to 1 to 2% ~~changes-of-change in~~ the regional albedo of snow and ice. This change in snow albedo was estimated to cause a radiative effect of $+1.7 \text{ W m}^{-2}$ in April–May (average value for the Arctic north of 60° 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.

7 Summary and conclusions

In this study, we investigate an aerosol transport event from Europe to the European Arctic using measurements as well as regional chemical-transport model simulations for the first time. Specifically, an event involving long-range transport of biomass burning and anthropogenic aerosols from Europe to the Arctic in April 2008 is studied using the regional model WRF-Chem (8 bin MOSAIC aerosol scheme), to quantify impacts on aerosol

concentrations and resulting direct shortwave radiative effects in the Scandinavian Arctic. Modeled aerosols are evaluated against ground-based observations from the EMEP network in European source regions, and using POLARCAT-France aircraft measurements aloft in the European Arctic. The model reproduces background $\text{PM}_{2.5}$ levels at EMEP ground based stations in Europe (NMB = -0.9%) and in Arctic polluted air masses measured by the ATR42 aircraft (NMB = $+8.8\%$). Comparison with EMEP measurements shows that the model overestimates concentrations of particulate NO_3^- (NMB = $+107\%$) and NH_4^+ (NMB = $+53\%$) in source regions, probably because of overestimated NH_3 emissions, and may underestimate OC. Good agreement is found between simulated SO_4^{2-} and EMEP measurements (NMB = -0.6%).

The model indicates that European biomass burning and anthropogenic emissions both had a significant influence on total aerosol mass concentrations ($> 20\%$ of total $\text{PM}_{2.5}$) during portions of the POLARCAT-France spring campaign measurements analyzed in this study. Plumes influenced by biomass burning sources in the model are also found to be significantly influenced by anthropogenic emissions. These modeled mixed plumes contain elevated organic carbon and black carbon concentrations. They originated in Eastern Europe and Western Russia, and followed low altitude (below 2 km) transport pathways into the Arctic. Significant wet scavenging is predicted in the model during transport over Finland, reducing PM_{10} levels by 55%. Modeled high-altitude anthropogenic plumes, originating in central Europe, were rapidly uplifted (from 1 to 6 km in less than 24 h) by warm conveyor belt circulations over Poland and the North Sea. The model also predicts significant wet scavenging during transport of these anthropogenic plumes (PM_{10} reduced by 74%). Evaluation of the model against in-situ measurements and LIDAR profiles below the aircraft shows that the model correctly represents the average vertical distribution of aerosols during this European transport event, as well as the magnitude of the aerosol optical properties. However, this comparison suggests that the model is under representing the rate of aerosol growth processes, especially condensation, which has the largest impact on the older mixed plumes (3 to 5 days old).

The model is used to investigate the average vertical structure of aerosol enhancements from European anthropogenic and biomass burning emissions in the Scandinavian Arctic. Anthropogenic emissions are shown to influence aerosols at both low (~ 1.5 km) and higher altitudes (~ 4.5 km), while biomass burning emissions influence aerosols between these altitudes (2.5 to 3 km). BC and SO_4^- aerosol concentrations are proportionally more enhanced at lower altitudes, including at the surface.

This transport event brought elevated aerosol concentrations north of the Arctic Circle for a rather short period of 4 days, from 8 to 12 April 2008. Due to the location of the polar front, these European aerosols did not mix significantly with local Arctic air further north. However, this event is particularly interesting because of the extensive seasonal snow cover present in Northern Scandinavia during this period. We show that the event had a significant local atmospheric warming effect over snow and ice surfaces. The average 96 h TOA direct and semi-direct shortwave radiative effect from this event over snow and sea ice is found to be $+0.58 \text{ W m}^{-2}$ north of 60° N . At solar noon, in regions significantly influenced by European aerosols, larger warming is predicted, $+3.3 \text{ W m}^{-2}$ (TOA direct and semi-direct radiative effects) over the Scandinavian and Finnish snow cover north of 60° N . This result is of the same order of magnitude as values previously reported for aerosols in the western Arctic (Brock et al., 2011; Quinn et al., 2007).

These radiative effect values do not include the impacts of cloud/aerosol interactions, which could be ~~quite large and compensate the~~ significant due to the extensive cloud cover in Northern Scandinavia during this transport event. The indirect effect could offset the warming effect of European aerosols over snow and ice-covered surfaces ~~-This we have shown here.~~ Moreover, the indirect aerosol effect is still uncertain, especially in the Arctic, and further work is needed to estimate its magnitude. During POLARCAT-France, the ATR-42 aircraft also sampled an intense Asian plume that was not investigated in this study, which focuses on European aerosols. The contribution of Asian sources to Arctic pollution is an active area of research, and the POLARCAT-France dataset, as well as the other POLARCAT datasets, could be the basis of a focused study on the transport of such plumes to the Arctic.

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Table 1. Parameterizations and options used for the WRF-Chem simulations.

Atmospheric process	WRF-Chem option
Planetary Boundary Layer	MYJ (Janjic et al., 1994)
Surface layer	Monin–Obukhov Janjic Eta scheme (Janjic et al., 1994)
Land surface	Unified Noah land-surface model (Chen and Dudhia, 2001)
Microphysics	Morrison (Morrison, Thompson and Tatarskii, 2009)
SW radiation	Goddard (Chou and Suarez, 1999)
LW radiation	RRTM (Mlawer et al., 1997)
Photolysis	Fast-J (Wild et al., 2000)
Cumulus parameterization	Grell-3 (Grell and Devenyi, 2002)
Gas phase chemistry	CBM-Z (Zaveri and Peters, 1999)
Aerosol model	MOSAIC 8 bins (Zaveri et al., 2008)

Table 2. Modeled PM_{2.5} aerosol composition by source type along POLARCAT-France spring flights. BC, OC and SS are black carbon, organic carbon, and sea salt, respectively.

Flight	Source type	BC (%)	OC (%)	SO ₄ ⁼ (%)	NH ₄ ⁺ (%)	NO ₃ ⁻ (%)	SS (%)
9 Apr 2008	Anthro.	2.5	7.0	24.1	20.6	40.2	5.6
	Mixed fires + anthro.	3.2	12.6	35.0	20.1	26.0	3.2
10 Apr 2008	Anthro.	2.3	5.5	21.7	20.9	42.4	7.3
11 Apr 2008	Anthro.	2.7	8.7	34.4	19.5	27.3	7.4
	Mixed fires + anthro.	2.8	11.9	33.9	19.4	28.5	3.4

Table 3. 4 day average shortwave direct and semi-direct effect (DSRE) at top of atmosphere (TOA) north of 60° N, over regions significantly influenced by European pollution (> 50% of total PM_{2.5} column due to in-domain anthropogenic and biomass burning emissions).

Type of land surface	DSRE at TOA (W m ⁻²)
Snow and Ice cover > 90 %	+0.58
Ocean	-1.52
All	-0.98

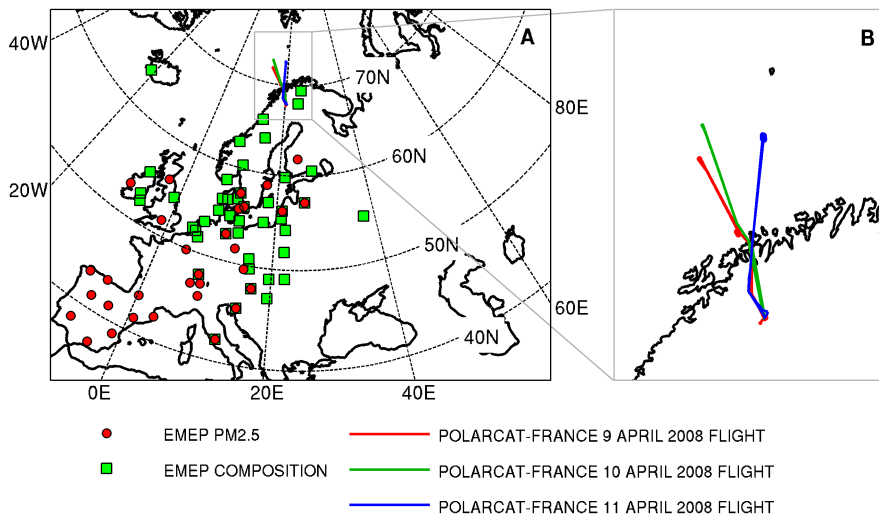


Figure 1. (a) WRF-Chem domain including the location of ground based EMEP measurement stations used for this study. Stations measuring PM_{2.5} are marked by a red circle, and stations measuring aerosol composition are marked by a green square. Stations with both measurements are indicated with both symbols. The POLARCAT-France spring flight tracks are shown in red, green and blue, with a zoom in over the flight region shown in (b).

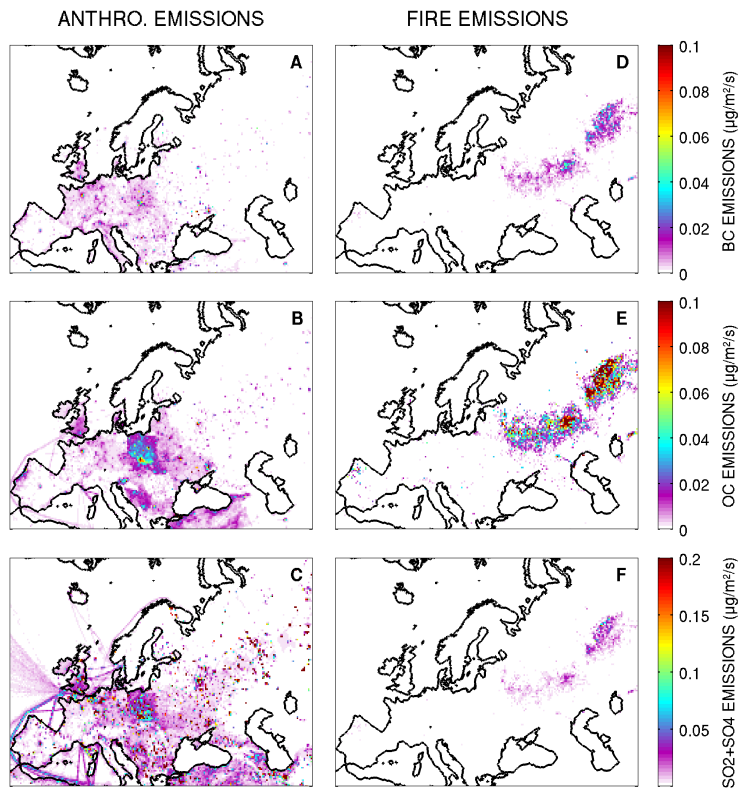


Figure 2. Averaged emissions within the model domain during the simulation period (1 April 2008–12 April 2008) due to anthropogenic activities (HTAP v2) and biomass burning (FINN v1). Anthropogenic BC, OC, and SO₂ + SO₄ emissions are shown in (a–c) and biomass burning BC, OC and SO₂ + SO₄ emissions are shown in (d–f).

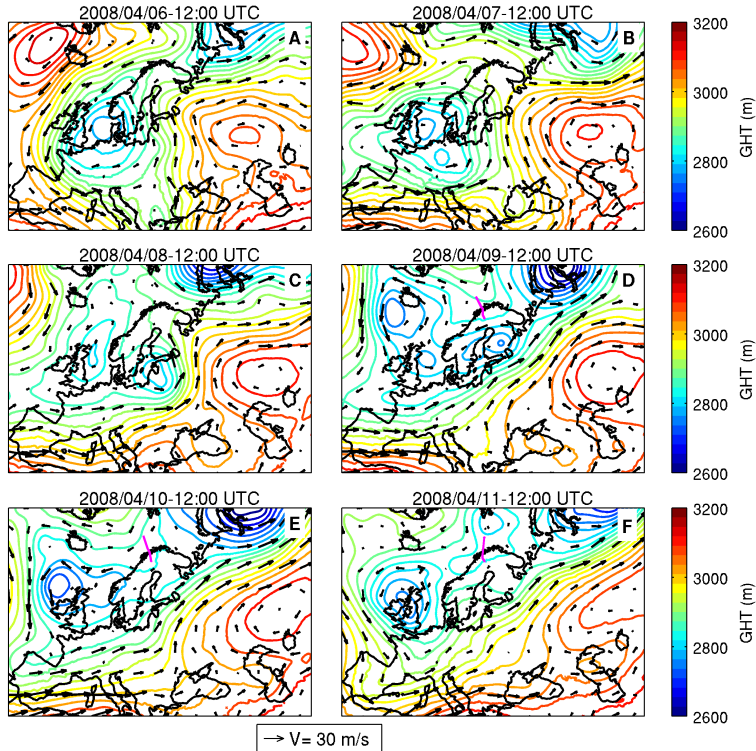


Figure 3. Meteorological situation simulated by WRF-Chem during the POLARCAT-France spring campaign period, represented by the 700 hPa geopotential height (contour lines) and 700 hPa wind vectors (30 m s^{-1} vector given for scale) on 6–11 April 2008 (12:00 UTC). The POLARCAT-France flight tracks on 9, 10, and 11 April 2008 are indicated in magenta.

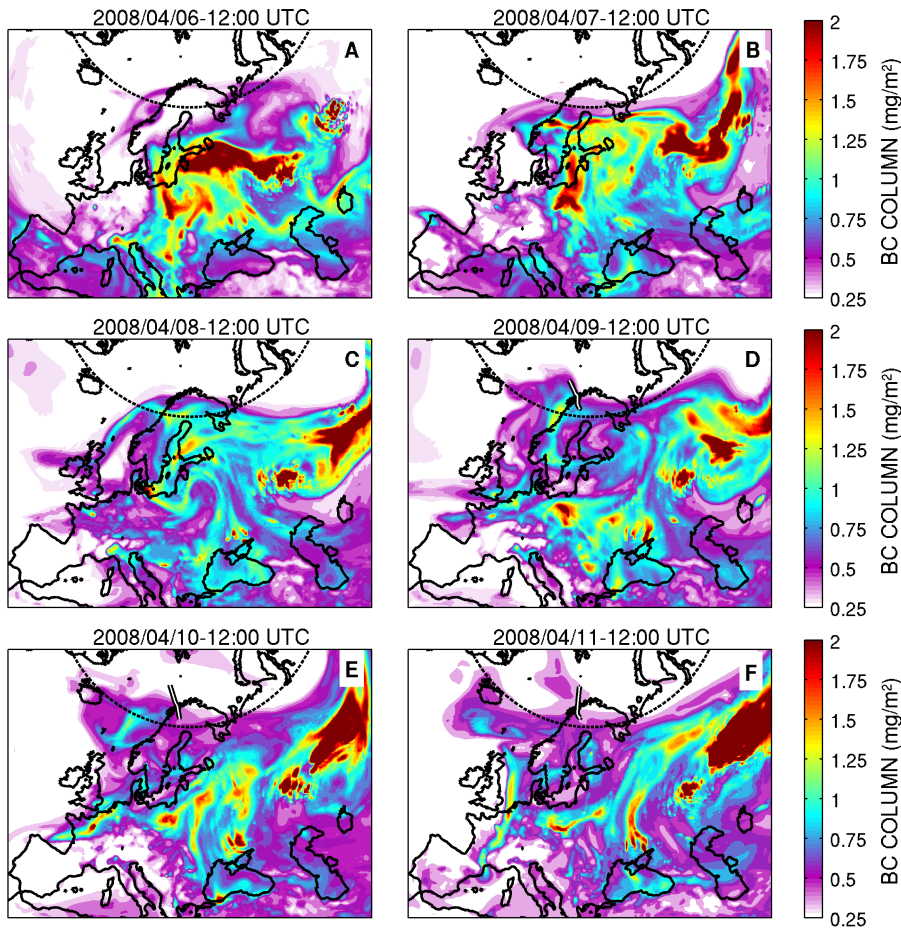


Figure 4. Simulated BC column on 6–11 April 2008 (12:00 UTC). POLARCAT-France flight tracks are indicated in white, with a black border.

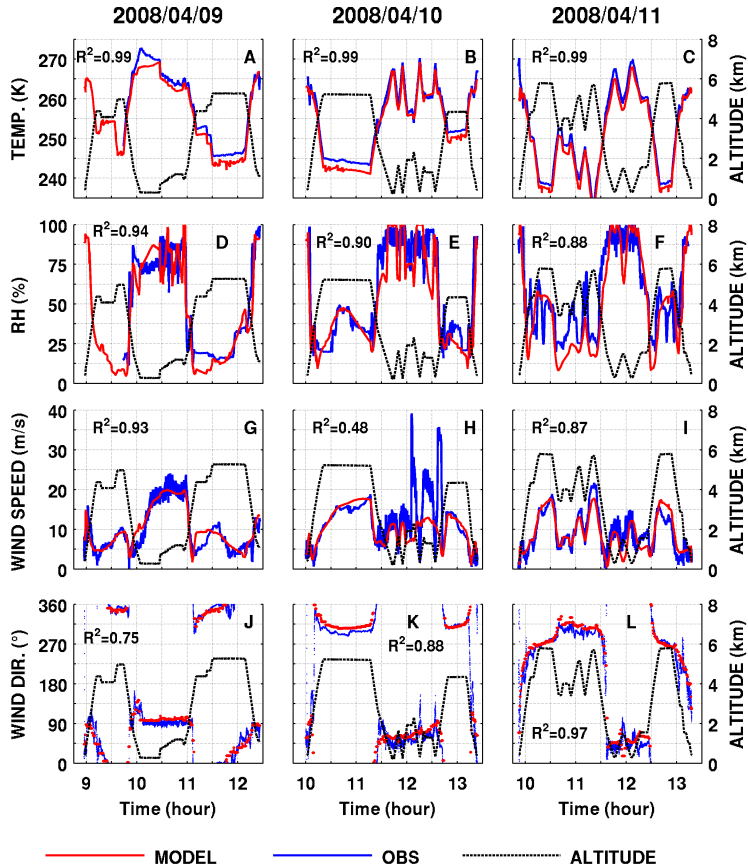


Figure 5. Time series of modeled (red) and measured (blue) (a–c) temperature, (d–f) relative humidity, (g–i) wind speed, and (j–l) wind direction extracted along the POLARCAT-France flight tracks. The corresponding aircraft altitude is shown in black.

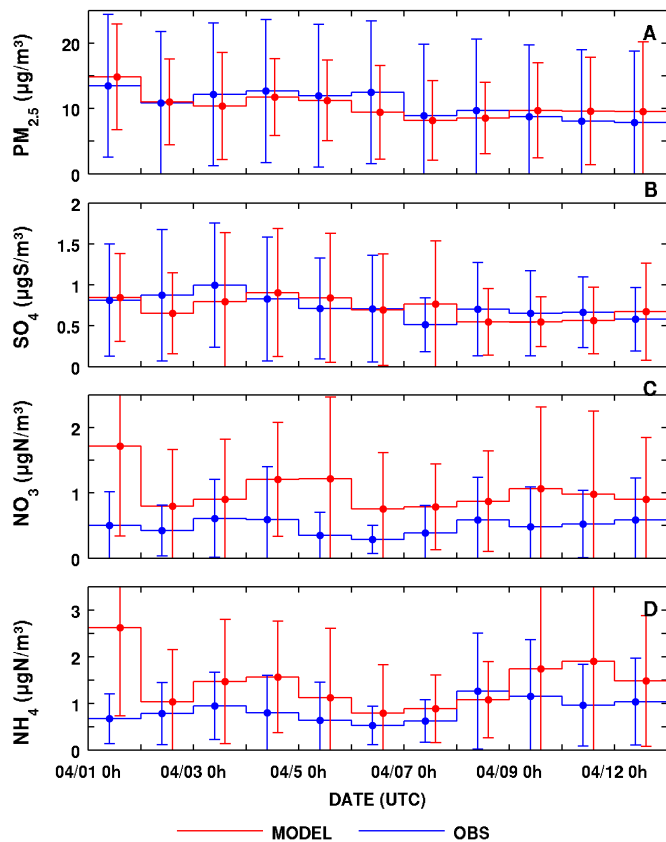


Figure 6. Daily mean aerosol mass measured at EMEP stations within the domain (in blue) and WRF-Chem aerosol mass extracted at the position of the stations (in red) for **(a)** PM_{2.5}, **(b)** sulfate aerosol, **(c)** nitrate aerosol, **(d)** ammonium aerosol. The standard deviation between stations is indicated by the error bars.

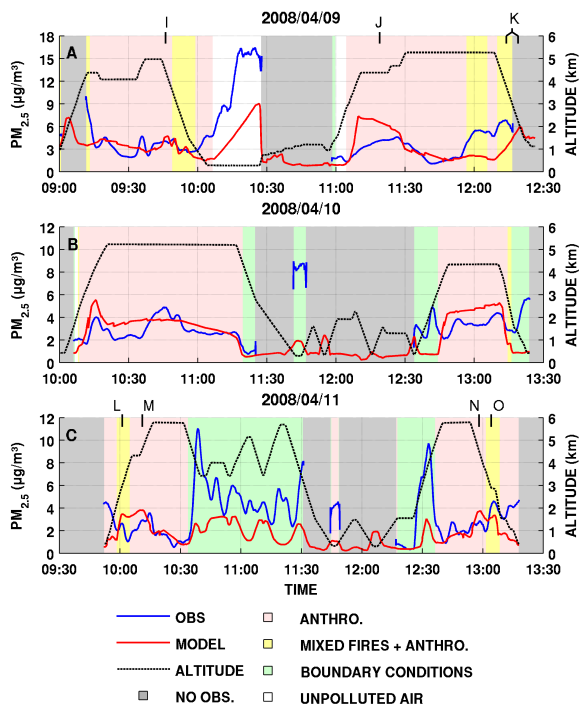


Figure 7. Time series of PM_{2.5} measured during POLARCAT-France (blue) and modeled (red) with the aircraft altitude indicated in black for the three POLARCAT-France flights on (a) 9 April 2008, (b) 10 April 2008 and (c) 11 April 2008. Grey shading indicates times when no measurements are available. Colors indicate when PM_{2.5} was significantly influenced (> 20% of PM_{2.5}) by source: green = air entering the domain from the northern boundary conditions, pink = anthropogenic emissions within the domain, yellow = fire emissions within the domain, [white = unpolluted air \(free of recent pollution sources\)](#). Letter labels indicate anthropogenic (I, J, M, N) and mixed anthropogenic/fire (K, L, O) plumes investigated further.

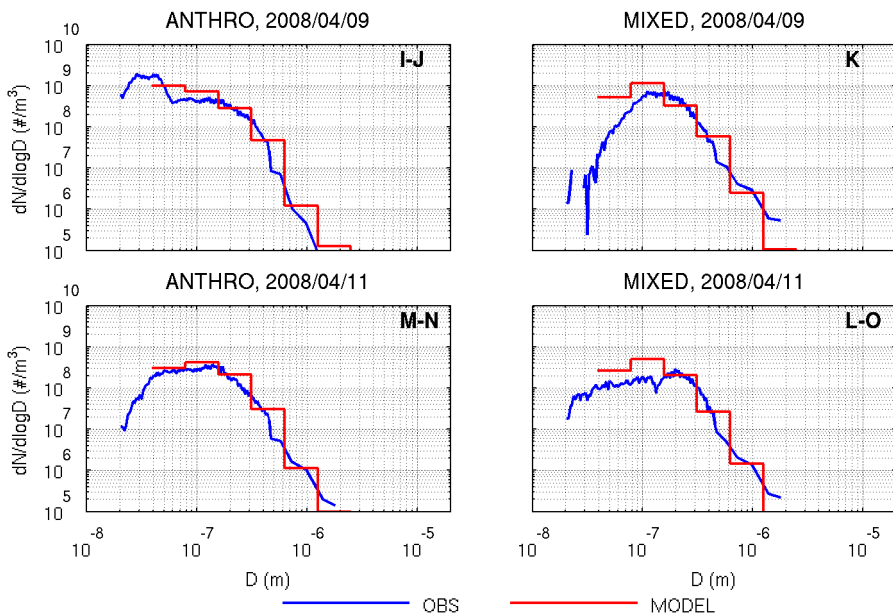


Figure 8. Modeled (red) and measured (blue) number size distributions of plumes labeled (I–O) in Fig. 7, influenced by (I, J, M, N) European anthropogenic and (K, L, O) mixed European anthropogenic and fire emissions. Modeled and observed size distributions corresponding to two consecutive samplings of the same plume during the same flight (I–J, M–N, L–O) were averaged together.

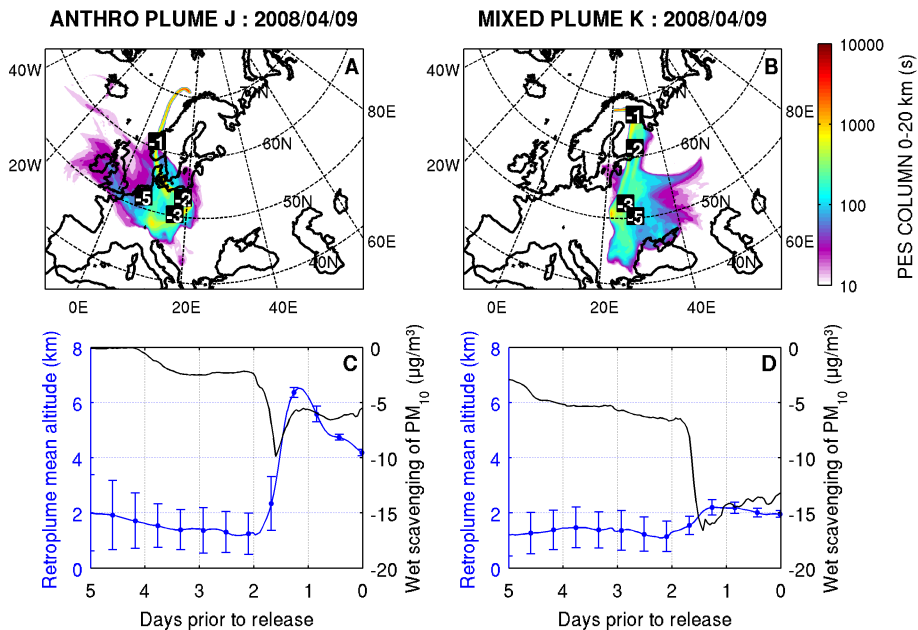


Figure 9. Backward mode FLEXPART-WRF column integrated PES (a and b) showing typical transport pathways for an anthropogenic plume (left, plume J, originating on 9 April 2008 at 11:19 UTC on the POLARCAT flight track) and a mixed anthropogenic/biomass-burning plume (right, plume K, originating on 9 April 2008 at 12:19 UTC on the flight track). (c and d) show each plume's mean altitude with RMS error bars showing vertical dispersion (blue) and the difference between the CTL PM_{10} and the NOWETSCAV PM_{10} along transport, indicating wet scavenging events (black).

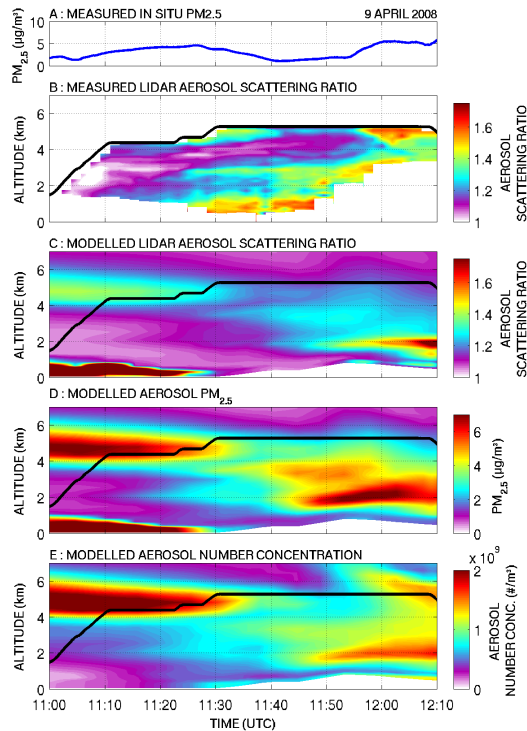


Figure 10. (a) PM_{2.5} measured in situ during the last part of the 9 April 2008 flight, (b) LIDAR 532 nm pseudo backscatter ratio measured at nadir during the same portion of the flight (altitude in black, white areas represent topography or cloudy areas where no aerosol data is available), (c) simulated WRF-Chem LIDAR 532 nm pseudo backscatter ratio, (d) modeled PM_{2.5} cross-section at the same position, (e) modeled aerosol number concentration cross-section at the same position.

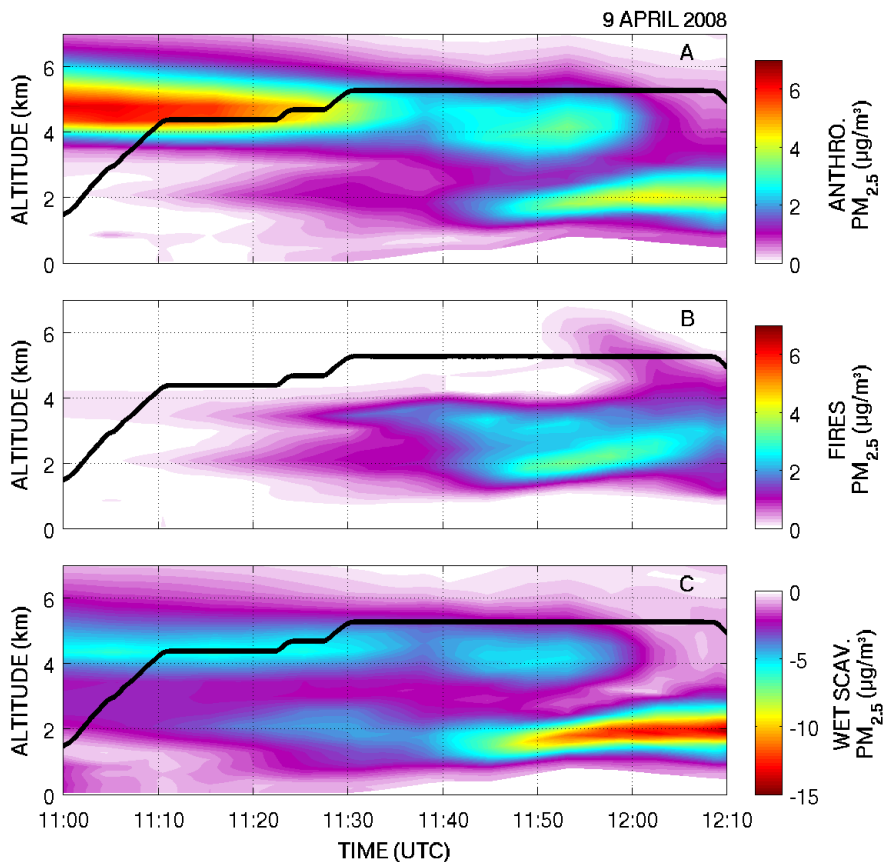


Figure 11. Modeled aerosol cross-sections along the flight track (plane altitude in black), showing the sensitivity of the modeled $PM_{2.5}$ to (a) anthropogenic emissions, (b) fire emissions, and (c) wet scavenging.

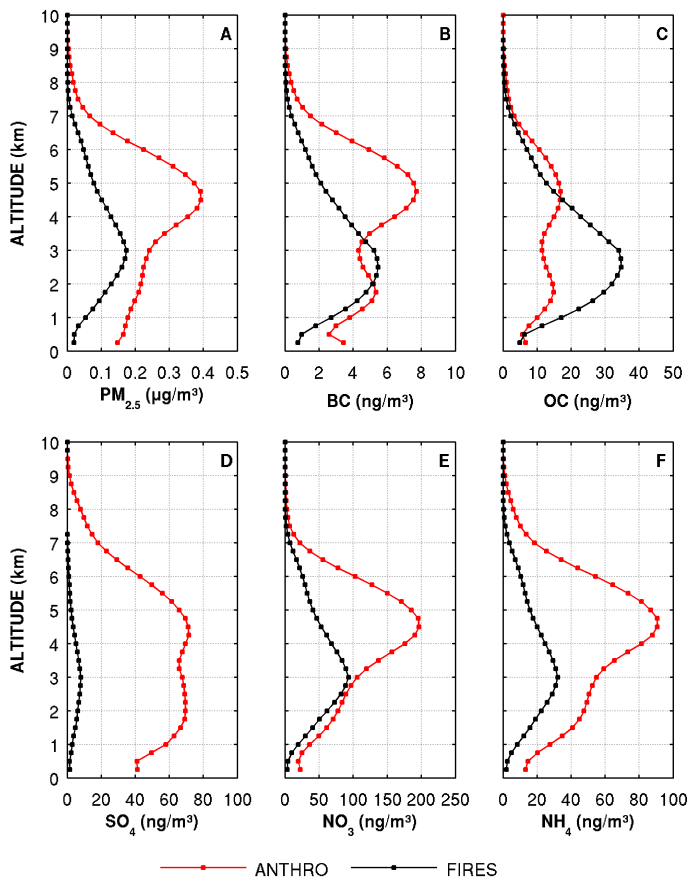


Figure 12. Modeled vertical profiles of enhancements in **(a)** $\text{PM}_{2.5}$, **(b)** BC, **(c)** OC, **(d)** $\text{SO}_4^{=}$, **(e)** NO_3^- and **(f)** NH_4^+ $\text{PM}_{2.5}$, due to anthropogenic (red) and fire (black) emissions within the WRF-Chem model domain, averaged in the Arctic (latitude $> 66.6^\circ \text{N}$) and over the period from 00:00 UTC 8 April 2008 to 00:00 UTC 12 April 2008.

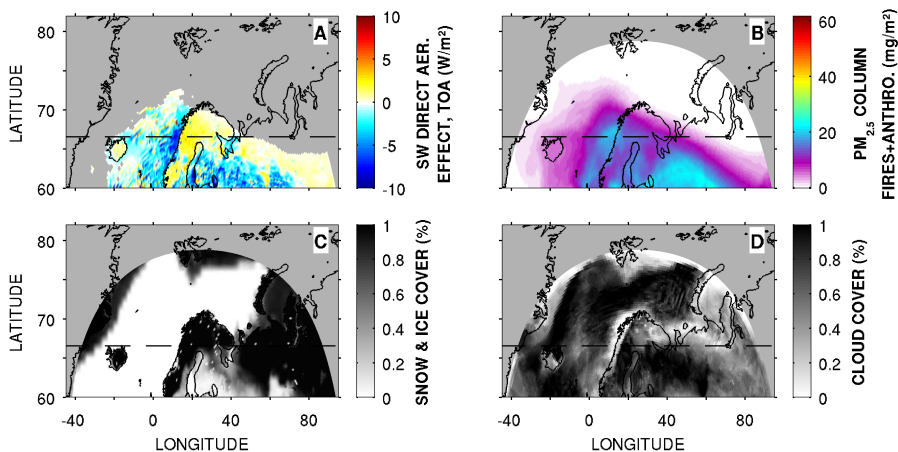


Figure 13. Model averages over the period from 00:00 UTC 8 April 2008 to 00:00 UTC 12 April 2008 of the: **(a)** aerosol direct and semi-direct radiative effect (DSRE), at Top Of Atmosphere (TOA), in regions significantly affected by in-domain anthropogenic and fire emissions, **(b)** PM_{2.5} column sensitivity to anthropogenic and biomass burning emissions, **(c)** fractional snow and sea ice cover, **(d)** fractional cloud cover. In panel **(a)**, regions not significantly affected by in-domain emissions are masked in gray. In panels **(b–d)**, regions outside of the WRF-Chem domain are masked in gray. The Arctic Circle is indicated by a dashed line.