

Interactive comment on “Origin of elemental carbon in snow from Western Siberia and northwestern European Russia during winter–spring 2014, 2015 and 2016” by Nikolaos Evangeliou et al.

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General Comments: I have no show-stopping issues with the analysis presented or the paper. The authors present a fairly comprehensive analysis of EC from snow samples collected across northern Russia, compare them to modeled values, and do a source apportionment analysis using FLEXPART in a new mode that allows running back trajectories that track deposited mass, rather than ambient atmospheric concentrations. They also compare their EC concentrations to those from other measurements around the Arctic, and they test modeled EC against these concentrations from other studies.

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The data set and analysis presented are useful and the paper should be published once the issues raised below are addressed.

Response: We would like to acknowledge the reviewer for his very constructive comments on some issues that we had not taken into account before. We believe we have addressed all of his comments.

Specific Comments: Very minor editing for English would be good. (e.g. “a component of the fine particulate matter” “a component of fine particulate matter”; “further tried to futher analyze”; “TRA and DOM contributed double to snow BC sampled at low latitudes”. . .).

Response: Corrected.

The sampling dates varied from early Feb to late April. When the samples were collected could influence the results in two ways that are not sufficiently discussed: 1) Biomass burning (wildfires) in northern Eurasia can become significant in March to April. The source apportionment (Fig 2) shows a very small role of wildfires, but there is some influence in some of the northern samples in 2015 and a significant role in one of the samples in 2016. It might be useful to indicate in Fig 2 (perhaps above each bar?) what date the samples were collected. 2) It seems possible there might have been some surface melting of the snow before sampling. If this is the case, surface concentrations could be elevated due to consolidation of BC at the snow surface, rather than due to increased deposition. Was there any effort made to determine whether the snow might have experienced melt at some point prior to being sampled? Either way, this should be noted.

Response: We agree with the reviewer that these reasons could potentially change concentrations of snow EC a lot.

As for biomass burning, we found a small contribution to snow BC in the majority of the samples. However, we believe that this is more or less expected simply because sam-

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pling took place in spring-time and it has been previously show using several different approaches and datasets that the hot season of biomass burning in Eurasia is rather summer (see Hao et al. doi:10.5194/gmd-9-4461-2016). We also wanted to put dates on Figure 2. However, due to lack of enough space to put dates in a comprehensive way, we decided to put coordinates and place all the meta-data of the samples in a separate Table that is placed in the Supplements of this article (Table S1).

As regards to the possibility of collection of melting snow, the sapling campaigns in these 3 years were designed such as that the sampling included only fresh snow and NOT melting snow. I now make in more clear in the beginning of section 2.1.

Hegg et al. found that biomass burning constituted a significant fraction of BC in snow from their northern Russia samples, in contrast to what you found here (i.e. see Fig 2). Hegg et al. could not distinguish between wildfire emission and domestic wood-burning emissions, so one possible explanation is that a significant fraction of the DOM (domestic burning) category in this study is wood burning. This would bring the source attribution of Hegg et al. and that given here in better agreement. It would be very useful if you could state what is included in the DOM emissions category; whether or not for this region a significant fraction of the DOM emissions are from wood burning; and to compare your source apportionment results to that of Hegg et al.

Response: Hegg et al. have used a completely different approach to address contribution of different sources to snow BC using a chemical analysis combined with Positive Matrix Factorization. On the contrary, we used a Langrangian Particle Dispersion Model combined with the most updated gridded emissions from ECLIPSEv5 (see methodology in section 2). This means that we use a preset portion of each of the sources that is already know and well documented in ECLIPSE website (see: http://www.iiasa.ac.at/web/home/research/researchPrograms/air/Global_emissions.html) and in Klimont et al. paper (doi: 10.5194/acp-17-8681-2017). In the aforementioned paper, section 3.1 describes all the approaches that used to produce what in the paper we call as DOM sector. Again, to be clear, all the differ-

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ent emission sectors used is ECLIPSE and they were used to generate Figure 2 are constant gridded sources from ECLIPSE. Furthermore, Hegg et al. has treated with his PMF model lots of chemical measurements from 36 samples in high latitudes. If you look at the Supplementary information of his article (http://pubs.acs.org/doi/suppl/10.1021/es803623f/suppl_file/es803623f_si_001.pdf), only 8 samples were collected from the vicinity of Russia (the rest were from Greenland, N. Pole, and N. America) and from completely different regions as we did. Therefore, we do not see how I would compare 2 different things both in terms of methodological and spatiotemporal manner.

Pg 2, lines 42-44. “Modelled BC was in good agreement ($R = 0.53 - 0.83$) with measured EC. However, a systematic region-specific model underestimation was found.” The wording here needs editing. First, R is an measure of correlation, not agreement. R could be 1.0, but if the two differ by a factor of 2 there is hardly “good agreement”. Second, an R of 0.53 means R-squared of 0.28, which is not a very high correlation coefficient. I would say they were moderately correlated, and the measured values were higher than the modeled values (by, e.g. “on average, XX%”).

Response: Corrected.

Pg. 9, lines 246-250: Same comment as made above re: the text in the Abstract around “good agreement”, and confounding “correlation” and “agreement”. As discussed in the text that follows, there was often significant bias in the modeled values relative to the measured values!

Response: Corrected.

Pgs. 12-13 and Figure S2 discussion of comparison of FLEXPART and Doherty et al. (2010) results: First, Figure S2 would be more useful if it showed the locations of the samples compared in a map and then the actual comparison in an x-y correlation plot. Trying to compare the two maps as given is not very useful, given the large range in concentrations. In an x-y plot, locations in different regions could be given different

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symbols, corresponding to the regional comparisons (e.g. Canadian Arctic, Western Siberia) as discussed in the text. Second, again, the text significantly over-states the level of agreement. In this case R is 0.24 ($R^2 < 0.06$ – i.e. the model only captures <6% of measured variability - !), and there is a 50% bias in the concentrations, on average.

Response: We partly agree with the reviewer in this comment. However, when plotting the data on a x-y correlation plot we end up with Figure 1.

This figure basically shows nothing and no comparison can really be done. This is expected, because the data range within 2 orders of magnitude (0.3 to >400 ng/g) and hence the only way to show them in an x-y plot is to use logarithmic axes like in Figure 2. Here, the modeled results are more centered towards the 1:1 line, but still the plot does not say the overall truth, because the axes are in a log scale. Furthermore, in the last figure, we know nothing about where exactly the model fails to predict measurements, which is not the case when plotting the data on a map.

Because the figure refers to data that were used for supporting validation only and it is only shown in the Supplementary information of this article, we would like to keep the figure as it is. If the reviewer still insists, we are willing to change it in a next step of the reviewing process. The sentences on correlations and agreements have been corrected.

Pg. 13, line 397: Again, R of 0.63 (R^2 of 0.29) is not “quite high”

Response: Corrected.

Pg. 13-14: Hegg et al. (2010, ACP) presents a source attribution of the BC in Arctic Canada snow measured by Doherty et al. (2010). It would be good to incorporate these results in the discussion here. Not doing so seems like an omission.

Response: I have tried to incorporate some of the main results at the end of the first paragraph in section 4.1, although I do not see how this study is related to what we try to do here, given that we use the data for validation only and NOT for interpretation.,

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Pg. 17, lines 521-522: “The model captured levels of BC quite effectively despite the large variation in measured concentrations.” Again, I disagree with this very optimistic statement of the results of the comparison.

Response: Corrected.

Smaller comments/corrections:

Pg 2, lines 45 and 47: The use of $>-100\%$ and $<-100\%$ is a bit ambiguous. “ $>-100\%$ ” could be read as more than a factor of 2 difference, and “ $<-100\%$ ” as less than a factor of 2 difference. I’d suggest rewording for better clarity.

Response: We agree and have corrected this part.

Pg. 3 lines 73-75: “Sea ice has a much higher albedo ($\approx 0.5-0.7$) compared to the surrounding ocean (≈ 0.06), thus BC deposited on sea ice reduces the heat uptake of the ocean.” I understand what you’re trying to say here, but as written it’s not accurate: BC deposited on ice does not reduce the heat uptake of the ocean – the presence of sea ice does. BC deposited on ice lowers its albedo, increases heat uptake by sea ice, accelerates its melt, and therefore decreases surface albedo both directly and indirectly.

Response: Corrected.

pg 4, lines 111-112: After discussing (correctly!) that BC/EC are operationally defined it’s stated that “In the present study, EC measurement data from three campaigns are compared to simulation results” – without stating what measurement method is used!

Response: Corrected.

Pg 5, lines 133: It is well known that quartz-fiber filters can have low and highly variable capture efficiency for particles in liquid samples. Was capture efficiency tested/measured? If not, at a minimum this potential source of bias needs to be acknowledged. Hopefully, some tests were done. (As an example, Hadley et al., 2008,

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Env Sci Tech found that to get high filter capture efficiency they had to run the samples through 3 stacked filters (Lij)

Response: We acknowledge that the collection efficiency of BC in liquid samples by quartz fiber filters can be less than 100%, as reported by Ogren et al. (1993) and Hadley et al. (2008). Differences in collection efficiencies between quartz fiber filters from different manufacturers, and even between batches, can be speculated. Unfortunately, no attempt to estimate the collection efficiency was performed in the present study, and estimating this based on previous studies is speculative. Thus, the results presented should be regarded as conservative estimates. We have included a sentence in the paper to account for this (see line 182-185).

Pg. 6. Line 161: I would reword “driven with 3-hourly” to “3-hour resolution”

Response: Modified.

Pg. 7, lines 201-203: “Assumed aerodynamic mean diameter and logarithmic standard deviation are used by FLEXPART’s dry deposition scheme, which is based on the resistance analogy. . .” The assumed size for BC (0.25 microns) is reasonable. However, the deposition rate should be driven by the size of the particles *containing* the BC. It is very unlikely that the BC in the atmosphere was externally mixed with other aerosol components; much more likely is that multiple components were internally mixed in larger particles. This would affect dry deposition rates based on resistance.

Response: BC particles in fresh exhaust are typically found in the 100 nm range or smaller and, in the urban environment, grow relatively quickly to sizes of about 200 nm (e.g., Ning et al., 2013). We agree that this occurs mainly via internal mixing with other types of aerosols. In remote areas, BC is mostly part of the internal aerosol mixture, with typical sizes of around 200 nm (see Freud et al., 2017, for Arctic size distributions). The wet diameters (which determine the physical behavior of the particles such as settling) will be larger than that.

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FLEXPART uses a single size distribution for BC aerosols and it does not account for particle growth. Therefore, a size distribution must be chosen that is representative for a broad range of conditions. Our size distribution is not representative for the external mixture of fresh BC particles (which are much smaller) but rather for the internal mixture of aerosols encountered in the Arctic and during most of the time BC resides in the atmosphere. It would not be appropriate to simulate the behavior of BC in fresh exhaust. Thus, while we totally agree with the reviewer about the mixing state of the BC particles, we think our settings are representative of this.

REFERENCES

Freud, E., Krejci, R., Tunved, P., Leaitch, R., Nguyen, Q. T., Massling, A., Skov, H., and Barrie, L.: Pan-Arctic aerosol number size distributions: seasonality and transport patterns, *Atmos. Chem. Phys.*, 17, 8101-8128, <https://doi.org/10.5194/acp-17-8101-2017>, 2017.

Ning, Z., Chan, K.L., Wong, K.C., Westerdahl, D., Mocnik, G., Zhou, J. H., Cheung, C.S.: Black carbon mass size distributions of diesel exhaust and urban aerosols measured using differential mobility analyzer in tandem with Aethalometer, *Atmos. Environ.*, 80, 31-40, 2013

Pg. 10, pg 287-289: Doherty et al. (2010) specifically measured BC in snow in northern Russia, including western Russia. It's odd not to note this, and to not compare your results directly with theirs from a similar region. Also it's odd to only state that concentrations were "up to 800ng/g", rather than discussing more representative results from their analysis.

Response: Done! We have added a short comparison in section 4.1 about BC and BC measured in samples from the Yamal peninsula. We only give an overview of the levels of concentrations as the information on the metadata is rather poor, the samples were not taken from exactly the same coordinates and they were also collected in different years.

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Pg. 26, Figure 1 caption: It might be good to remind the reader in the caption that the ECLIPSE emissions don't include wildfire emissions.

Response: Modified.

Pg. 27, Figure 2 caption: Some rewording/re-parsing of the (very long!!!) first sentence of this caption would make it much more readable. . .

Response: Modified.

Figure 1: Right-most panel, showing spatial distribution of EC concentrations. I found the color-scale used here not very intuitive. It might be better to go from, e.g., dark blue for low values to bright red for high values.

Response: If we do what the reviewer suggests we end up with Figure 3. The figure shows no color variation on the measured EC concentrations. This is because the majority of the measured concentrations were between 0-100 ng/g (blue) and only 3-4 samples above. Therefore, we would like to keep the same colorbar as before in order to show discrete colors for all samples.

Figures 3-5: I found the little red stars indicating sampling location difficult to find. I'd suggest making this symbol larger.

Response: Modified.

Interactive comment on Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2017-542>, 2017.

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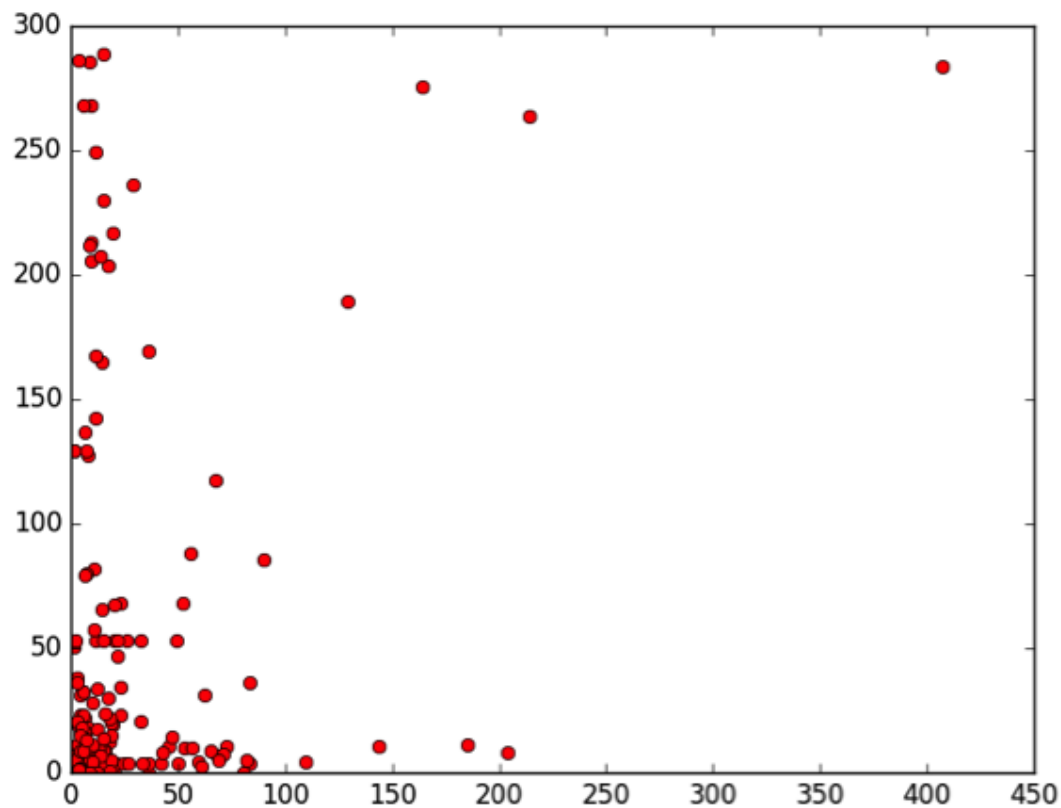


Fig. 1.

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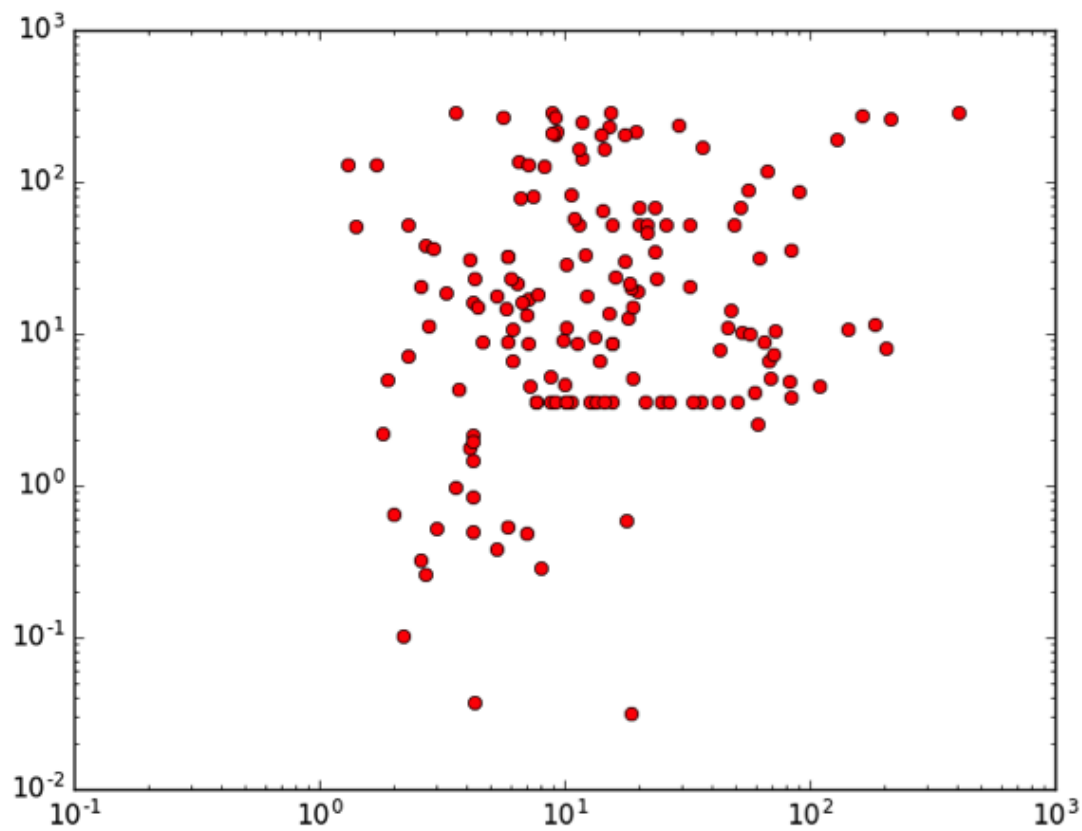


Fig. 2.

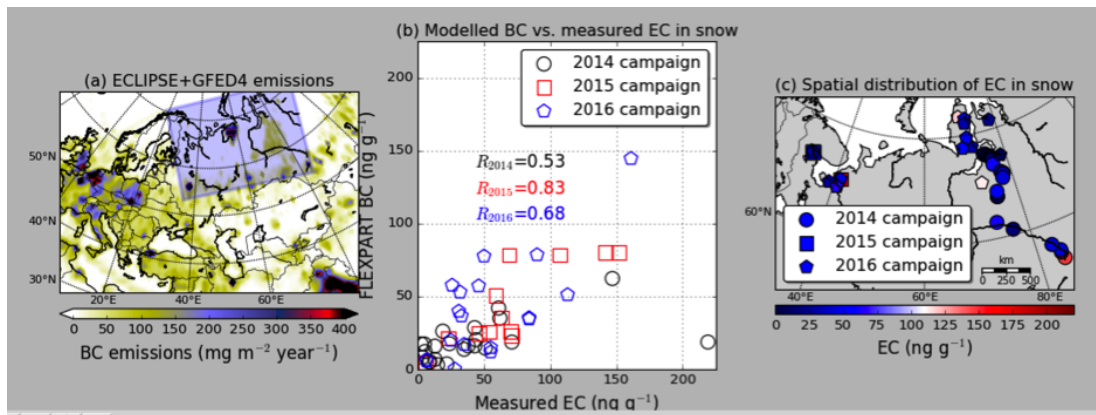


Fig. 3.

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