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RESPONSE TO Anonymous Referee #1

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.

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 sampling 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 Lagrangian 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 known 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 different 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 ($\delta IS' E \sim 0.53 - 0.83$) 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 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,.

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

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.

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., Leitch, 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.

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.

Fig. 1

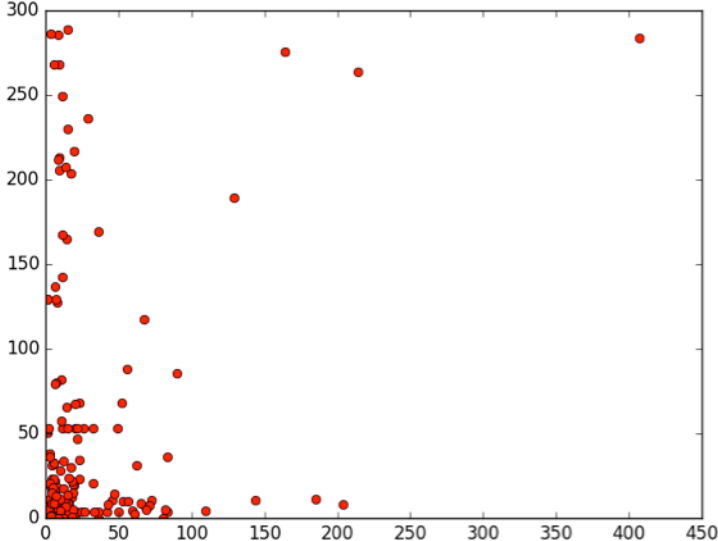


Fig. 2

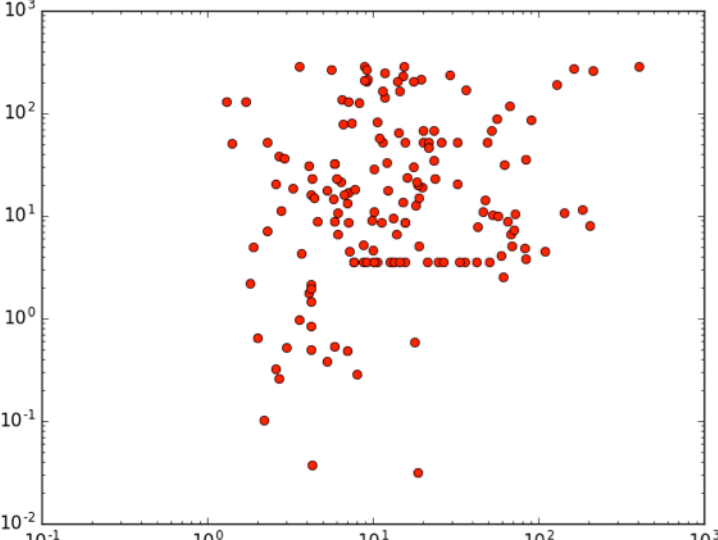
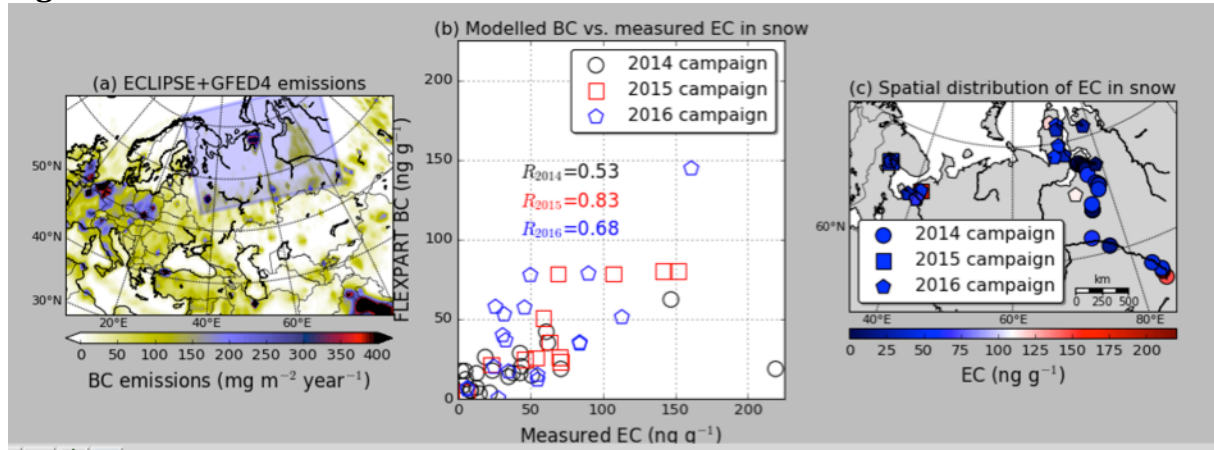


Fig. 3



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Origin of elemental carbon in snow from Western Siberia and northwestern European Russia during winter-spring 2014, 2015 and 2016 Author: Nikolaos Evangelidou et al.

General Comments

1. The manuscript present results of elemental carbon (EC) concentration in snow samples collected at various locations in the regions of Western Siberia and northwestern European Russia. It also presented output of LDMP FLEX- PART model which was used to identify the major sources which contributed to the BC concentrations measured in the snow samples.

Response: We appreciate reviewer for his thorough grammar and syntax editing. Indeed the manuscript reads much better now and it is clearer in sections that were not previously.

2. The manuscript needs adequate grammar editing. The construct and flow of some of the sentences need to be re-constructed. The use of opening phrasal nouns and adjectives are often out of place.

Response: We have followed reviewer's suggestions. We appreciate reviewer's help for this.

3. A major deficiency of the manuscript is the labelling of the individual plots in the Figures. Figures 1 - 7 should be labelled (a), (b), (c), (d) and so on as appropriate. This will make your discussion of the figure easier.

Response: Corrected.

4. The sentences in lines 41 and 52 are contradicting each other.

Response: Corrected.

5. Section 3 (Results) of the manuscript presented both results and discussion of the various analysis rather than the results. Detailed discussions of the results should be in section 4 (Discussion) along with the cross validation of the model and model deviation.

Response: Generally, we are not keen on presenting measurements or modeling results with numbers only. We also like to trigger deeper on what data represent, otherwise we have a rather boring manuscript.

In this sense, we present the results of the snow concentrations of BC in section 3 and the simulations of the LPDM FLEXPART in the same section. In discussions' section, we have a more general discussion of our results, perform an extra validation of the new

feature of our model and also try to find potential patterns of areas that our model fails or succeeds to capture measured concentrations.

We believe that this structure is appropriate for the presentation of this study. However, if the reviewer insists to restructure the whole manuscript, we would need further, more specific, instructions.

6. Under sample collection, it is necessary to highlight the number of samples collected at each site and the total number for each year. Also, provide a separate figure of the sampling sites preferably a map. When making reference to the sampling site, you refer the readers to Figure 1 which did not show the sampling sites explicitly.

Response: The total number of samples analysed per year is now shown in lines 139-142.

We do not really understand what else is needed at this point. Figure 1 (a) shows a map of Europe and a shaded blue area, which is the sampling location. Then we zoom in the highlighted region and present the sampling points in Figure 1 (c). In Figure 1 (c), we use 3 different markers corresponding to each different year and also show different colors that correspond to BC concentrations in a colormap.

7. For the concentration of EC in snow (section 3.1), you could report the percentile (upper and lower) instead of the standard deviation.

Response: The number of samples that we measured during the three campaigns in 2014, 2015 and 2016 was 23, 11 and 20, respectively. We think that the number of samples is very low to present percentiles. On the other hand, presenting minimum and maximum ranges and medians with standard deviations certainly gives an overview of the concentrations level, which is also shown in Fig. 1 (c) and in Fig. 2.

8. Some of the data compressed into section 3.1 could be better understood by the reader if they are presented in tabular form.

Response: Corrected. Please see Table S2 in the Supplementary Information.

9. For the cross validation (section 4.1), it will be better to state explicitly that you used FLEXPART to simulate BC concentration for Doherty and Macdonald's dataset.

Response: Corrected. Please see in line 458 and 525, respectively.

10. You have used different reference format for the manuscript and supplementary materials.

Response: Corrected.

11. In line 142, what is the performance compared with?

Response: Usually, accredited laboratories are obliged to participate in intercomparison exercises. This is a common procedure and there are several references about this. Thus we frequently measure EC in filter samples using the TOA technique for such intercomparisons.

12. In section 2.3, what do you mean by carbonate (CO₂-3)-carbon? Do you mean carbonate (CO₂-3)?

Response: It is the carbonate content of CO₃²⁻ which is the issue when it interferes with the OC and EC during TOA, not the oxygen content. The term is commonly used when addressing such issues. We would like to keep this term in the manuscript.

Specific Comments:

1. Line 38: Why did you refer to the recently developed algorithm as feature? I think it should be recently developed algorithm or routine.

Response: We understand that the reviewer is not experienced with the Lagrangian Particle Dispersion Model (LPDM) FLEXPART. So far, the model (or the algorithm is you like) could track atmospheric concentrations back in time. In the development we have done (see Eckhardt et al., 2017), in the same model framework (FLEXPART), we have added the possibility for the user to be able to simulate wet and dry deposition back in time. This is simply done by changing one parameter (details can be seen in Eckhardt et al. paper). This is the reason that we refer to this development as feature to the already existing model framework FLEXPARTv10.

2. Line 39: backwards should be backward

Response: Corrected.

3. Line 57: most strongly should be strongest. Delete 'the'. That part of the sentence should read "component of atmospheric aerosol."

Response: Corrected.

4. Line 65: should read "BC is important on a global perspective because of its
..."

Response: Corrected.

5. Lines 65-66: provide a reference for the opening sentence.

Response: References are given in the next sentences.

6. Line 66: should read “As a component of fine particulate matter”

Response: Corrected.

7. Line 69: should read “. it absorbs radiation and accelerates melting of the ice.

Response: Corrected.

8. Lines 91-93: The references cited here are not properly cited. The last part after the unnecessary full stop should not be in a bracket.

Response: Corrected.

9. Line 101: are major sources of what in the area?

Response: Corrected.

10. Lines 104 -105: The references in the bracket should be preceded by ‘for example’ since the references are just examples of articles that have used EC and BC.

Response: Corrected.

11. Lines 107 – 110: The statement “consequently, BCthe world” added no substantial meaning to the discussion here. Hence, I suggest you expunge it.

Response: We disagree. This sentence tells a lot about BC and points to the long-term discussion between experimentalists and modellers for the right use of terms BC, eBC, rBC etc. We would like to keep it as it is.

12. Line 110: should read “.....BC should be used quantitatively”

Response: Corrected.

13. The statement “In the present study,” should start a new paragraph.

Response: Corrected.

14. Line 119: re-cast the statement beginning from “near the port”. The near near in the last part of the statement makes it ambiguous.

Response: Corrected.

15. Line 120: Is Kindo Peninsula in Arkhangelsk or Arkhangelsk is a sampling site on its own?

Response: Here, we clearly point to three different areas. We do not understand why the reviewer thinks that Kindo P. is in Arkhangelsk.

16. Line 121: should read “. Pollution levels in these areas have been partly attributed to urban and gas flaring sources.”

Response: Edited to better much reviewer’s suggestions.

17. Line 125: should read “. to minimise the direct influence from”

Response: Corrected.

18. Line 126: should read “. information about sample collection such”

Response: Corrected.

19. Line 127: should read “. and the depth at which snow was sampled”

Response: Corrected.

20. Line 129: should read “Sampling was perform with a metal-free technique using pre-cleaned”

Response: Corrected.

21. Line 130: should read “. polyethylene bags which had been”

Response: Corrected.

22. Line 131: should read “. 1M HCl and rinsed with abundant deionised ultrapure water in the”

Response: Corrected.

23. Line 133: should read “ filtered through 47 mm quartz fibre filters. The filters were dried at 60-70 oC”

Response: Corrected but kept specifications of the filters in the manuscript.

24. Line 138-139: should read “Ele- mental carbon content of the filters were measured (TOA) using the sunset laboratory”

Response: Corrected.

25. Line 142: should read “Performance of the OC/EC instrument is regularly”

Response: Corrected.

26. Line 143-144: Recast leaving out the slash af- ter (EMEP).

Response: Corrected.

27. Line 148-149: should read “The carbonate content of filtrate on the filters was measured by TOA after thermal-oxidative”

Response: See major comments number 12.

28. Line 150: should read “A punch of 1.5 cm²”

Response: Corrected.

29. Line 152-154: Re-cast this sentence. Do you mean section 2.2 or chapter 2.2?

Response: Corrected. “Section” fits much better here.

30. Line 156 and 157: ‘evolves’ should be ‘evolved’

Response: Corrected.

31. Line 158: should read “Applying this correction, EC values were”

Response: Corrected.

32. Line 160: Give the full meaning of LPDM at first use.

Response: The full meaning of LPDM is given at the last paragraph of Introduction.

33. Lines 163-165: Re-cast to read “The ECMWF data has 137 vertical data and a horizontal resolution of 1 x 1 for 2014 and 2015 simulation, and 0.5 x 0.5 for 2016”

Response: Corrected.

34. Line 188: mass per unit area you mean

Response: Corrected.

35. Could you re-cast this sentence?

Response: Perhaps the reviewer has forgotten adding the line where the sentence to be re-casted is.

36. Line 198-200: What are the rationale/references for these assumptions? Any similar assumption in literature?

Response: Of course. I re-write the response to a relevant comment from reviewer 1: 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.

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.

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

37. Line 214: this should be Figure 1(c).

Response: Corrected.

38. Line 216: Like I stated in the general comment, you could report the 25th and 75th percentile or 10th and 90th percentile.

Response: I think that the number of samples that we measured in not sufficient for that. We have maintained this presentation of concentrations (see major comment).

39. Line 221: should read “.....the snow samples for 2014, EC con- centrations”

Response: Corrected.

40. Line 228: should read “.....(on the White sea coast) showed high”

Response: Corrected.

41. Line 232-240: Re-cast the five sentences in these lines.

Response: We have try to edit these sentences, but we do not know towards which direction as the comment is not very specific..

42. Line 239: should read “Tomsk and Yamal, EC concentration was highly”

Response: Corrected.

43. Line 244: Should read “.....measured EC concentrations in the snow samples”

Response: Corrected.

44. Line 246: A scatter plot of what? Figure 1 should be Figure 1(b).

Response: Corrected.

45. Line 247: should read “..... agreement and good correlation

Response: Edited.

46. Line 258: The sentence “The MFB of the was -42%” is somehow isolated. What inference can be drawn from the fact that MFB is -42%.

Response: Corrected.

47. Line 264: should read “For 2016, FB values show another set of underestimation.

Response: Corrected.

48. Line 266: 12 out of 19 what? Samples?

Response: Corrected. Yes, we meant samples.

49. Line 266: Should read “19 samples. For the remaining 7 samples, the model

Response: Corrected.

50. Line 267 should read “..... The root mean square error

Response: Corrected.

51. Line 268-269: Please, re-frame this sentence. The sentence, as it stands, is ambiguous. I guess it should read “The RMSE is frequently used to measure

Response: Corrected.

52. Line 273-275: the sentence is muddled up. What exactly do you want the reader to infer from the two short sentences?

Response: Corrected.

53. Line 276-277: should read “..... reported that the maximum BC concentration measured

Response: Corrected.

54. Line 283: should read “.....Stockholm with a population of about 2 million.

Response: Corrected.

55. Line 287: What do you mean by one order of magnitude?

Response: Corrected.

56. Line 290: should read “. Macdonald et al., (2017) reported BC concentrations ranging from For the samples collected near

Response: Corrected.

57. Line 295: should read “.....ECLIPSE emissions dataset” The word ‘account’ does not work here. Please, choose a different word.

Response: Corrected.

58. Line 297- 298: should read “..... gas flaring (FLR) while biomass burning”

Response: Corrected.

59. Line 310-311: the list of cities in the bracket is just too long. Include only the important cities and move the bracket to immediate after ‘major Russian cities on line 309.

Response: Corrected.

60. Line 313-316: re-cast this sentence to reflect what you want the reader to understand from the sentence,

Response: Corrected.

61. Line 319: should read “(6%) (see Figure 2).....”

Response: Corrected.

62. Line 320-321: Are these two sources new? Where they not there in 2014?

Response: No, they are not new at all. It is simply the fact that samples collected from different regions are usually influenced by different sources.

63. Line 325-326: should read “..... Peninsula whereas FLR emissions were very low due to the long distance of flaring emission sources from the sampling point.”

Response: Corrected.

64. Line 327-328: should read “. . . also affected BC concentration in snow in northwestern”

Response: Corrected.

65. Line 329: should read “ releases in Russia, the miscalculation and their impact in”

Response: Corrected.

66. Line 331-332: should read “.BB emissions, originating mostly from eastern Europe, contributed about”

Response: Corrected.

67. Line 336: should read “. . . Yamal, DOM, FLR TRA contributed, on the average, 31%, 29% and 27%, respectively (see Figure 2(c)).”

Response: Corrected.

68. Line 341: it should be Figure 5(b) if you effect the comment on labelling of individual figures in the plot as suggested in the general comment section

Response: Corrected.

69. Line 353-359: Re-phrase the sentences on these lines stating what exactly you did will the data from Doherty and Macdonald as well as the reasons for the cross validation.

Response: Corrected.

70. Line 374-375: should read" Similar to our finding for the new Russian mea- surements, the model output, with a MFB of -51%, tends to underestimate deposition."

Response: Corrected.

71. Line 383-384: Are you referring to Doherty data here? If so, state that explicitly.

Response: It is now stated explicitly in the beginning of the paragraph (line 507).

72. Line 388: Expunge "Moreover" The sentence should read" Our model output was . . .with measured BC concentrations in"

Response: Corrected.

73. Line 392: 'research' should be 're- searcher'

Response: We believe not! It is a small population of research and military personnel.

74. Be explicit. Did you do a model run for the period for which Macdonald et al carried out measurements?

Response: Corrected. Please see line 545.

75. Line 401: underestimated what?

Response: Corrected.

76. Line 402: should read "Further analysis was carried out to adequately understand . . ."

Response: Corrected.

77. Line 404-408: Re-cast this complex sentence into 2 – 3 simple ones.

Response: Corrected.

78. Line 413: should read ".....Two hotspots were"

Response: Corrected.

79. Line 414: should read "... And an- other, of smaller intensity, in southeastern Asia."

Response: Corrected.

80. Line 415-417: The two simple sentences here are disjointed.

Response: Corrected.

81. Line 419: should read"... .. America in ECLIPSE. The Alert samples, for which the model strongly underestimated BC, the major sources".

Response: Corrected.

82. Line 421: Why is 7 ng g-1 not in percentage?

Response: Corrected.

83. Line 422: should read"... .. Alert air pollutant concentrations"

Response: Corrected.

84. Line 429: should read "It has been shown that average measured"

Response: Corrected.

85. Line 432: delete 'already.'

Response: Corrected.

86. Line 437: should read"... .. locations of fires that have been active in the last two months before the sample collection. The fire data were adopted from MODIS ..."

Response: "Active fires" is a very common product of MODIS and this is the reason that we want to keep this expression. The rest has been corrected according to the reviewer's suggestion.

87. Line 439: gas flaring facilities or gas flaring data?

Response: It is "gas flaring facilities" what we plot in Figure 7.

88. Line 443: How do you mean? Around gas flaring facilities?

Response: Khanty-Mansiysk region is known among scientists that study BC transport as one of the most important regions of gas-flaring emissions in the world. Yes, there are many facilities of this type in the area, and they can be easily seen from space (see VIIRS data in Fig. 1):

Circle shows Nenets-Komi and rectangle Khanty/Mansiysk regions.

89. Line 445: should read". According to a related study by Huang and Fu (2016),"

Response: Corrected.

90. Line 450: Which model are you refer- ring to here?

Response: Corrected.

91. Line 451-452: These cities/regions are not explicitly labelled in the plots. So that the reader can follow through with the discussions, it is better to include lon/lat of these cities/regions in a bracket. Could you do this for other locations in sim- ilar discussion throughout the manuscript at their first mention?

Response: These regions mentioned are well known among scientists that study BC. E are talking about the most important global sources of BC located inside the Polar Dome, which directly affect the Arctic. We believe that adding so much information in the figures will put a lot of pressure on how to make everything visible to the readers (cities, regions of interest, colors representing data, etc...).

92. Line 452: should be Figure 7(b)

Response: Corrected.

93. Line 458-459: should read". northwestern Russia, a region which includes Murmansk. Pollution level in Murmansk could be high due to"

Response: Corrected.

94. Line 462: You have referred to figure 7 severally but these cities are not explicitly shown in Figure 7.

Response: See response in comment 91. These regions are easily seen by the hotspots that are visible in Figure 7.

95. Line 467: should read" polluting sources identified in ECLIPSE dataset."

Response: Corrected.

96. Line 471: should read "to have originated mainly"

Response: Corrected.

97. Line 472-473: Re-cast this sentence. Insignificant? Negligible?

Response: Corrected.

98. Line 474: delete (Figure S5). It makes no contribution to the sentence.

Response: Moved below.

99. Line 475: should read" European Russia (Figure S5)"

Response: Corrected.

100. Line 476: should read"of the total contribution, which reflect the proximity of the sampling site to the main flaring facilities in Russia."

Response: Corrected.

101. Line 477-479: could you re-phrase this sentence?

Response: Corrected.

102. Line 480: delete Figure (S6). It adds nothing to the understanding of this sentence,

Response: Moved below.

103. Line 482: Delete 'Again' sub-categories should be 'categories.'

Response: Corrected.

104. Line 483-484: What could be responsible for the insignificant contribution of FLR at this sampling site? Is the site upwind of the flaring facilities? It would be interesting if you could put forward an argument for this insignificant contribution despite the closeness to the flaring facilities.

Response: Corrected.

105. Line 499: : should read" Russia in 2014, 2015 and 2016 EC concentration"

Response: Corrected.

106. Line 501-502: should read" to measured BC concentration in snow"

Response: Corrected.

107. Line 507: : should read" Russian emission as well as"

Response: Corrected.

108. Line 515: should read " emissions originating from highly"

Response: Corrected.

109. Line 525-526: should read" Considering the fact that similarobserved in samples collected in the area during other years, it is likely . . . of BC in this region show"

Response: Corrected.

110. Line 528: should read" . . . previously reported average measurements of BC concentrations in snow in Western"

Response: Corrected.

111. Line 529: delete 'on average'.

Response: Corrected.

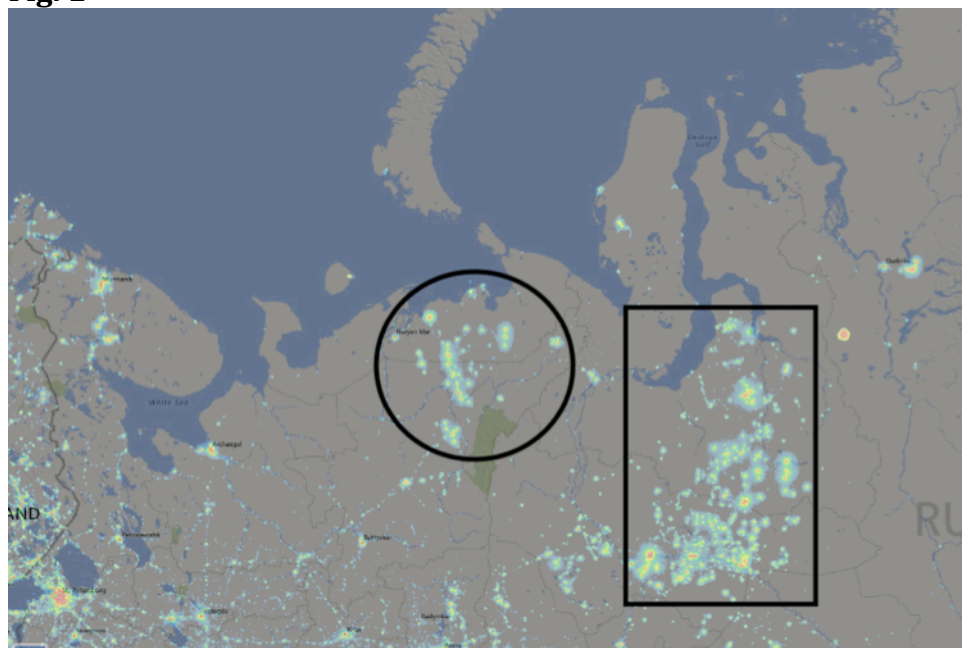
112. Line 584: Delete one of the 'doi'.

Response: Corrected.

113. Line 623-626: This reference is cited as 2016 in the manuscript (see line 207).

Response: Corrected.

Fig. 1



1 **Origin of elemental carbon in snow from Western Siberia**
2 **and northwestern European Russia during winter–spring**
3 **2014, 2015 and 2016**

4

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24

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28

29 **Abstract**

30 Short-lived climate forcers have been proven important both for the climate and human
31 health. In particular, black carbon (BC) is an important climate forcer both as an aerosol and
32 when deposited on snow and ice surface, because of its strong light absorption. This paper
33 presents measurements of elemental carbon (EC; a measurement-based definition of BC) in
34 snow collected from Western Siberia and northwestern European Russia during 2014, 2015
35 and 2016. The Russian Arctic is of great interest to the scientific community due to the large
36 uncertainty of emission sources there. We have determined the major contributing sources of
37 BC in snow in Western Siberia and northwestern European Russia using a Lagrangian
38 atmospheric transport model. For the first time, we use a recently developed feature that
39 calculates deposition in backward (so-called retroplume) simulations allowing estimation of
40 the specific locations of sources that contribute to the deposited mass.

41 EC concentrations in snow from Western Siberia and northwestern European Russia
42 were highly variable depending on the sampling location. Modelled BC and measured EC
43 were moderately correlated ($R = 0.53 - 0.83$) and a systematic region-specific model
44 underestimation was found. For EC sampled in northwestern European Russia the
45 underestimation by the model was smaller (fractional bias, $FB > -100\%$). In this region, the
46 major sources were transportation activities and domestic combustion in Finland. When
47 sampling shifted to Western Siberia, the model underestimation was more significant ($FB < -$
48 100%). There, the sources included emissions from gas flaring as a major contributor to snow
49 BC. The accuracy of the model calculations was also evaluated using two independent
50 datasets of BC measurements in snow covering the entire Arctic. The model reproduced snow
51 BC concentrations quite accurately, although small discrepancies occurred mainly for samples
52 collected in springtime. Nevertheless, EC concentrations in snow presented here are about
53 20% lower than previously reported ones in Western Siberia and northwestern European
54 Russia.

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62 1 Introduction

63 Black carbon (BC) is the strongest light-absorbing component of atmospheric aerosol
64 and is formed by the incomplete combustion of fossil fuels, biofuels, and biomass (Bond et
65 al., 2013). It is emitted directly into the atmosphere in the form of fine particles. BC is a major
66 component of “soot”, a complex light-absorbing mixture that also contains organic carbon
67 (OC) (Bond et al., 2004). Combustion sources emitting BC include open biomass burning
68 (forest, savanna, agricultural burning), residential biofuel combustion, diesel engines for
69 transportation or industrial use, industrial processes and power generation, or residential coal
70 combustion (Liu et al., 2011; Wang et al., 2011).

71 BC is important on a global perspective because of its impacts on human health and on
72 climate. As a component of fine particulate matter (PM2.5), it is associated with negative
73 health impacts, including premature mortality (Lelieveld et al., 2015; Turner et al., 2005). It
74 absorbs solar radiation, has a significant impact on cloud formation and, when deposited on
75 ice and snow, it accelerates ice melting (Hansen and Nazarenko, 2004). BC has a lifetime that
76 can be as long as 9–16 days (Bond et al., 2013). After its emission, BC can travel over long
77 distances (Forster et al., 2001; Stohl et al., 2006) and reach remote areas such as the Arctic.
78 Arctic land areas are covered by snow in winter and spring, while the Arctic Ocean is partly
79 covered by ice. Sea ice has a much higher albedo (≈ 0.5 – 0.7) compared to the surrounding
80 ocean (≈ 0.06), thus presence of sea ice reduces the heat uptake of the ocean. Snow has an
81 even higher albedo than sea ice and can reflect as much as 90% of the incoming solar
82 radiation (Brandt et al., 2005; Singh and Haritashya, 2011). BC deposited on ice lowers its
83 albedo, increases heat uptake by sea ice, accelerates its melting, and therefore decreases
84 surface albedo both directly and indirectly.

85 Hegg et al. (2009) reported that snow in the Arctic often contains BC at concentrations
86 between 1 and 30 ppb, which can cause a snow albedo reduction of 1–3% in fresh snow and
87 another 3–9% as snow ages and BC becomes more concentrated near the surface (Clarke and
88 Noone, 1985). This solar radiation reflecting capacity of snow insulates the sea ice, maintains
89 cold temperatures and delays ice melt in summertime. After the snow begins to melt and
90 because shallow melt ponds have an albedo of approximately 0.2 to 0.4, the surface albedo
91 drops to about 0.75 or even lower (0.15) as melt ponds grow and deepen (Singh and
92 Haritashya, 2011). These changes have been found to be important for the global energy

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101 balance (Flanner et al., 2007; Hansen and Nazarenko, 2004) and, if enhanced by BC,
102 contribute to climate warming (Warren and Wiscombe, 1980).

103 Although BC in Arctic snow and ice has been found to be important for the Earth's
104 climate (Flanner et al., 2007; Sand et al., 2015), its large-scale temporal and spatial
105 distributions and exact origin are still poorly quantified (AMAP, 2015). Efforts to determine
106 the concentrations of BC in snow across the Arctic were made by Clarke and Noone (1985),
107 Doherty et al. (2010, 2013), Forsström et al. (2013), Ingvander et al. (2013) and McConnell et
108 al. (2007). This paper presents measurements of Elemental Carbon (EC) concentrations in
109 snow samples collected in spring 2014, 2015 and 2016 in the Kindo Peninsula (White Sea,
110 Karelia), around Arkhangelsk in northwestern European Russia, and in Western Siberia. In
111 the latter area, gas flaring emissions are very important. Flaring emissions are highly
112 uncertain because both activity data and emission factors are largely lacking. According to the
113 Global Gas Flaring Reduction Partnership (GGFR)
114 (<http://www.worldbank.org/en/programs/gasflaringreduction>), nearly 50 billion m³ of gas are
115 flared in Russia annually. The Russian flaring emissions in the Nenets/Komi regions and in
116 Khanty-Mansiysk are the major sources in Western Siberia and northwestern European
117 Russia. It has been reported that gas flaring in Russia contributes about 42% to the annual
118 average BC surface concentrations in the Arctic (Stohl et al., 2013).

119 The use of the terms EC and BC has been the topic of several scientific papers (for
120 example, Andreae and Gelencsér, 2006; Bond et al., 2013; Petzold et al., 2013). Petzold et al.
121 (2013) defined BC as a substance with 5 properties (see Table 1 in Petzold et al., 2013), for
122 which no single measurement instrument exists that is sensitive to all of them at the same
123 time. Consequently, BC cannot uniquely be measured, although some of its properties can,
124 such as the absorption coefficient σ_{ap} and the elemental carbon (EC) concentration, both
125 commonly measured in atmospheric monitoring networks across the world. Hence, the term
126 BC should be used qualitatively,

127 In the present study, EC concentrations on ice from three campaigns measured with
128 Thermal-Optical Analysis (TOA) (see section 2.2) are compared to simulation results from
129 the Lagrangian particle dispersion model (LPDM) FLEXPART. The model is used here for
130 the first time to quantify the sources contributing to BC in snow in Russia adopting a special
131 feature that was developed recently.

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136 2 Methodology

137 2.1 Collection and storage of snow samples

138 Fresh snow samples were collected along a north–south transect between Tomsk and
139 the Yamal coast in February–March 2014 (23 samples), while in March 2015 sample
140 collection took place in the Kindo Peninsula and near the port of Arkhangelsk in the White
141 Sea (11 samples, Figure 1). Finally, in February–May 2016 samples were collected in the
142 Kindo Peninsula, in Arkhangelsk and between Tomsk and Yamal (20 samples). These areas
143 have been reported to receive pollution both from urban and gas flaring sources (Stohl et al.,
144 2013). For example, the gas flaring sources located in Yamal and Khanty-Mansiysk (Russia)
145 are in the main pathway along which sub-Arctic air masses travel to the Arctic (Stohl et al.,
146 2006). All sampling points were located more than 500 m away from roads to minimize the
147 direct influence from local traffic emissions. Information about sample collection such as the
148 location of sampling, the amount of snow collected and the depth at which snow was sampled
149 is reported in Table S1 and the sample locations are plotted in Figure 1.

150 Sampling was performed using a metal-free technique, using pre-cleaned plastic shovels
151 and single-use vinyl gloves. Samples were stored in polyethylene bags, which had been
152 thoroughly washed with 1 M HCl and rinsed with abundant deionised ultrapure water in the
153 laboratory prior to their use. After returning the samples to the laboratory, the snow was
154 allowed to melt at ambient temperature (18–20°C), and immediately filtered through quartz
155 47 mm fibre filters (2500QAT-UP, Pall, for samples collected in 2014 and QM-A Whatman for
156 samples collected in 2015 and 2016). The filters were dried at 60–70°C, wrapped in
157 aluminum foil and stored in a refrigerator. Quartz fiber filter collection efficiency of BC in
158 liquid samples can be less than 100% (Hadley et al., 2010; Ogren et al., 1983). To what extent
159 this has affected the levels reported in the present study is unknown. Thus the results
160 presented should be regarded as conservative estimates based on the assumption that some
161 BC might have been lost during filtration.

162 2.2 Elemental Carbon measurements by Thermal–Optical Analysis (TOA)

163 Elemental carbon (EC) content of the filters was measured at NILU’s laboratories by thermal-
164 optical analysis (TOA), using the Sunset laboratory OC/EC instrument operated according to
165 the EUSAAR-2 protocol (Cavalli et al., 2010). A 1.5 cm² punch was cut from the filtered
166 snow samples for the analysis. Transmission was used for organic carbon (OC) charring
167 correction. Performance of the OC/EC instrument’s is regularly intercompared as part of the

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187 joint European Monitoring and Evaluation Programme (EMEP) Aerosols, Clouds, and Trace
188 gases Research InfraStructure Network (ACTRIS) quality assurance and quality control effort
189 (Cavalli et al., 2015).

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190 2.3 Measurements of carbonate (CO_3^{2-})-carbon by Thermal-Optical Analysis 191 (TOA) following thermal-oxidative pre-treatment

192 The content of carbonate (CO_3^{2-})-carbon on the filters was measured by TOA,
193 following thermal-oxidative pretreatment based on the approach described by Jankowski et al.
194 (2008). A punch of 1.5 cm^2 from each filter was heated at $450\text{ }^\circ\text{C}$ for 2 hours in ambient air to
195 remove OC and EC, but not CO_3^{2-} -carbon. The filter punch was subjected to TOA
196 immediately (30 sec) after thermal-oxidative pre-treatment. The split time (between OC and
197 EC) obtained for each filter punch used to determine the filter samples' content of EC (section
198 2.2) was also used to apportion CO_3^{2-} -carbon to OC and/or EC. The influence of CO_3^{2-} -
199 carbon evolving as EC, was accounted for by the following equation:

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$$EC_{CO_3^{2-}}^{corr} = EC - EC_{CO_3^{2-}}$$

200 where $EC_{CO_3^{2-}}^{corr}$ is elemental carbon corrected for CO_3^{2-} -carbon that evolved as EC during
201 TOA, EC is elemental carbon and $EC_{CO_3^{2-}}$ is CO_3^{2-} -carbon that evolved as EC during TOA.
202 Applying this correction, EC values were 5-22% lower (see Supplementary Information).

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203 2.4 Emissions and modelling of black carbon

204 The concentrations of BC in snow were simulated with the LPDM FLEXPART version
205 10 (Stohl et al., 1998, 2005). The model was driven with operational meteorological wind
206 fields retrieved from the European Centre for Medium-Range Weather Forecasts (ECMWF)
207 of 3-hour (for the years 2014 and 2015) and hour (for the year 2016) temporal resolution. The
208 ECMWF data have 137 vertical levels and a horizontal resolution of $1^\circ \times 1^\circ$ for the 2014 and
209 2015 simulations and $0.5^\circ \times 0.5^\circ$ for the 2016.

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retrieved from the European Centre for Medium-
Range Weather Forecasts (ECMWF).

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210 The simulations were conducted in backwards time ("retroplume") mode, using a new
211 feature of FLEXPART to reconstruct wet and dry deposition with backward simulations
212 (Eckhardt et al., 2017). This new feature is an extension of the traditional possibility to
213 simulate atmospheric concentrations backward in time (Seibert and Frank, 2004; Stohl et al.,
214 2003). It is computationally efficient because it requires only two single tracer transport
215 simulations (one for wet deposition, one for dry deposition) for each measurement sample. To

229 reconstruct wet deposition amounts of BC, computational particles were released at altitudes
230 of 0 to 20 km at the locations where snow samples were taken, whereas to reconstruct dry
231 deposition, particles were released between the surface and 30 m at these locations. All
232 released particles represent a unity deposition amount, which was converted immediately (i.e.,
233 upon release of a particle) to atmospheric concentrations using the deposition intensity as
234 characterized either by dry deposition velocity or scavenging rate (for further details, see
235 Eckhardt et al., 2017). The concentrations were subsequently treated as in normal
236 “concentration mode” backward tracking (Seibert and Frank, 2004) to establish source-
237 receptor relationships between the emissions and deposition amounts. The termination time of
238 the particle release was the time at which the snow sample was collected, whereas the
239 beginning time was set as the time when the ECMWF precipitation at the sampling site,
240 accumulated backward in time, was equal to the water equivalent of the snow sample, up to
241 the specified sampling depth.

242 The model output consists of a spatially gridded sensitivity of the BC deposition at the
243 sampling location (receptor) to the BC emissions, equivalent to the backwards time mode
244 output for concentrations (Seibert and Frank, 2004; Stohl et al., 2003). BC deposition at the
245 snow sampling point can be computed (in mass per unit area) by multiplying the emission
246 sensitivity in the lowest model layer (the footprint emission sensitivity) with gridded
247 emissions from a BC emission inventory and integrating over the grid. The deposited BC can
248 be easily converted to BC snow concentration by taking into account the water equivalent
249 depth of the snow from ECMWF (in mm). In the present study, the ECLIPSE (Evaluating the
250 CLimate and Air Quality ImPacts of ShortlivEd Pollutants) version 5 emission inventory
251 (Klimont et al., 2016; Stohl et al., 2015) was used
252 (http://www.iiasa.ac.at/web/home/research/researchPrograms/air/Global_emissions.html).

253 The total emissions of BC from ECLIPSE in the areas of study are shown in [Figure 1](#), (left
254 panel).

255 BC was assumed to have a density of 2 g m^{-3} in our simulations and a logarithmic size
256 distribution with an aerodynamic mean diameter of $0.25 \text{ }\mu\text{m}$ and a logarithmic standard
257 deviation of 0.3. Each computational particle released in FLEXPART represents an aerosol
258 population with a lognormal size distribution (see Stohl et al., 2005). Assumed aerodynamic
259 mean diameter and logarithmic standard deviation are used by FLEXPART’s dry deposition
260 scheme, which is based on the resistance analogy (Slinn 1982), and they are consistent with
261 those used in other transport models (see Evangeliou et al., 2016; Shiraiwa et al., 2008).

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264 Below-cloud scavenging was determined based on the precipitation rate taken from ECMWF.
265 The in-cloud scavenging was based on cloud liquid water and ice content, precipitation rate
266 and cloud depth from ECMWF (Grythe et al., 2017). The FLEXPART user manual (available
267 from <http://www.flexpart.eu>) provides more information. All modelling results for this
268 sampling campaign can be viewed interactively at the URL
269 http://niflheim.nilu.no/NikolaosPY/SnowBC_141516.py.

270 3 Results

271 3.1 Elemental Carbon concentrations measured in snow

272 The spatial distribution of EC measured in snow samples from northwestern European
273 Russia and Western Siberia is shown in [Figure 1\(c\)](#) for each of the campaigns (2014, 2015
274 and 2016). There was large spatial variability in the distribution of EC in snow in 2014
275 ranging from 3 to 219 ng g⁻¹, with a median (\pm standard deviation) of 23 \pm 50 ng g⁻¹. The
276 highest EC concentrations in 2014 were observed in Western Siberia near Tomsk (147 to 219
277 ng g⁻¹). FLEXPART emission sensitivities for these samples showed that the air was coming
278 from the north and the east (see in http://niflheim.nilu.no/NikolaosPY/SnowBC_141516.py).
279 This explains the high concentrations of EC, as most of the anthropogenic BC sources are
280 located in these regions. In the rest of the snow samples [for 2014](#), EC concentrations between
281 4 and 170 ng g⁻¹ were observed. High concentrations were observed near the Ob River
282 coinciding with air masses arriving mainly from Europe. During the 2015 field campaign, EC
283 concentrations were the highest near Arkhangelsk (175 ng g⁻¹), for which FLEXPART
284 showed that the air was coming from nearby areas
285 (http://niflheim.nilu.no/NikolaosPY/SnowBC_141516.py). Therefore, it is likely that the
286 samples were affected by direct emissions from the city or the port of Arkhangelsk. During
287 the same campaign, snow samples collected in the Kindo peninsula ([on the White Sea coast](#))
288 showed high variability in EC concentrations (range: 46 – 152 ng g⁻¹, median=70 \pm 37 ng g⁻¹).
289 According to FLEXPART emission sensitivities, air masses were transported to Kindo
290 peninsula from central and southern Europe driven by an anticyclone over Scandinavia
291 (http://niflheim.nilu.no/NikolaosPY/SnowBC_141516.py). Finally, [for the](#) snow samples
292 collected outside Arkhangelsk, at the Kindo peninsula, [and](#) close to the Yamal Peninsula in
293 Western Siberia [in 2016, EC concentrations ranged between](#) 7–161 ng g⁻¹ (median: 40 \pm 39 ng
294 g⁻¹). [Outside](#) Arkhangelsk, EC concentrations varied widely from 31 to 161 ng g⁻¹ with a
295 median concentration in this region of 61 \pm 45 ng g⁻¹. This is far below the 175 ng g⁻¹ observed

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306 in 2015, although there was only one sample collected in that year. In the Kindo Peninsula,
307 EC was relatively constant in 2016 ranging between 25 and 35 ng g⁻¹ (median = 28±4 ng g⁻¹),
308 which is more than 60% lower compared with the 2015 values (median = 70±37 ng g⁻¹).
309 Finally, between Tomsk and Yamal, EC concentration was highly variable (7 – 119 ng g⁻¹)
310 due to the different EC sources affecting snow (median = 50±38 ng g⁻¹). For instance, it is
311 expected that gas flaring affects snow close to Yamal, while snow collected in the south
312 (Tomsk) is likely influenced by sources in Europe or local urban emissions. Nevertheless, the
313 highest concentrations (>100 ng g⁻¹) were observed north of 68°N, in the Yamal Peninsula.

314 We compared the measured EC concentrations in the snow samples with those
315 calculated by FLEXPART. For this, the emission sensitivities were multiplied with the total
316 emission fluxes from ECLIPSE (section 2.4). A scatter plot of modelled and measured snow
317 concentrations is presented in Figure 1, (b). The results show a good correlation between
318 modelled BC and measured EC concentrations for the 2015 and 2016 campaigns ($R_{2015} =$
319 0.83 and $R_{2016} = 0.68$, $p - value < 0.05$), but weaker correlation for 2014 ($R_{2014} = 0.53$,
320 $p - value < 0.05$). For further validation, the fractional bias (FB) of each individual sample
321 was calculated together with the mean fractional bias (MFB) for observed EC and modelled
322 BC for the 2014, 2015 and 2016 sampling campaigns as follows:

$$FB = \frac{C_m - C_o}{(C_m + C_o)/2} \times 100\% \text{ and } MFB = \frac{1}{N} \sum_{i=1}^N \frac{C_m - C_o}{(C_m + C_o)/2} \times 100\%$$

323 where C_m and C_o are the modelled BC and measured EC concentrations and N is the total
324 number of observations for each year. The FB for individual samples is shown in Figure S 1.
325 FB is a useful model performance indicator because it is symmetric and gives equal weight to
326 underestimations and overestimations (it takes values between -200% and 200%). It is used
327 here to show the locations where modelled BC concentrations in snow over- or underestimate
328 observations (see Figure S 1). The MFB of the model for the 2014 snow measurements was -
329 42%, which shows that the model underestimated observations. In total, the model
330 underestimated concentrations for 17 out of 23 samples with FB values ranging from -168%
331 to -30%, whereas for the rest (six samples) FB values ranged between 20% and 148%
332 (median: -56%±81%) (Figure S 1). In 2015, the MFB of the model was -48% (median: -
333 56%±32%), where 11 out of 12 values were underestimated by the model showing FB values
334 that ranged between -101% and -7% (one FB value was found to be 12%). For 2016, FB
335 values of the simulated concentrations of BC in snow show another set of underestimation

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345 (median: $-13\pm 73\%$) varying from -198% to -0.3% for 12 out of 19 samples. For the
346 remaining seven samples, the model predicted higher concentrations compared with
347 observations (10% to 75%) (Figure S 1). The root mean square error (RMSE) was computed,
348 which is frequently used to measure differences between values predicted by a model and the
349 values actually observed. RMSE values were estimated to be quite high, between 37 and 49
350 ng g^{-1} , due to the large variation of the observed EC concentrations.

351 The levels of EC in snow presented here are relatively high compared to previously
352 reported concentrations in the Arctic. Apart from Aamaas et al. (2011) who measured
353 maximum EC concentration in snow close to the airport of Svalbard of more than 1000 ng g^{-1} ,
354 most of the reported levels of EC in the relevant literature are close to our findings. For
355 instance, Ruppel et al. (2014) found that EC concentrations have been increasing up to 103 ng
356 g^{-1} since 1970 in Svalbard. McConnell et al. (2007) reported that the BC concentrations
357 measured at the D4 ice-core site in Greenland were 10 ng g^{-1} , at maximum, which most likely
358 originated from biomass burning in the conifer-rich boreal forest of the Eastern and Northern
359 United States and Canada. Forsström et al. (2013) reported concentrations as high as 88 ng g^{-1}
360 in Scandinavia, and lower ones at higher latitudes ($11\text{--}14 \text{ ng g}^{-1}$ in Svalbard, $7\text{--}42 \text{ ng g}^{-1}$ in
361 the Fram Strait, and 9 ng g^{-1} in Barrow). Svensson et al. (2013) collected snow samples from
362 Tyresta National Park and Pallas-Yllästunturi National Park in Sweden. Tyresta is a relatively
363 polluted site located circa 25 km from the city centre of Stockholm with a population of about
364 2 million people. Yllästunturi National Park is located in Arctic Finland and a clean site with
365 no major city influencing the local and regional air. The concentration of EC in Pallas-
366 Yllästunturi was between 0 and 140 ng g^{-1} , while in Tyresta the BC concentrations were up to
367 more than 7 times higher ($53\text{--}810 \text{ ng g}^{-1}$). Furthermore, Doherty et al. (2010) in the most
368 complete dataset for the Arctic snow and ice BC reported highly variable concentrations (up
369 to 800 ng g^{-1}) for five consecutive years (2005–2009). Finally, in the most recent dataset for
370 snow BC_2 , Macdonald et al. (2017) reported BC_2 concentrations ranging from 0.3 to 15 ng g^{-1}
371 were reported for the samples collected near the Alert observatory (see section 4.1).

372 3.2 Sources and origin of BC

373 We further analysed the model output in order to calculate relevant contributions from
374 various BC source types to BC concentrations in snow (for method description, see section
375 2.4). ECLIPSE emissions include waste burning (WST), industrial combustion and processing
376 (IND), surface transportation (TRA), power plants, energy conversion, and extraction (ENE),

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395 residential and commercial combustion (DOM), gas flaring (FLR), while biomass burning
396 (BB) emissions were adopted from the Global Fire Emissions Database, Version 4
397 (GFEDv4.1) (Giglio et al., 2013). The results are depicted in Figure 2 for the sampling
398 campaigns of 2014, 2015 and 2016 in Western Siberia and North-Western European Russia,
399 sorted from the northernmost to the southernmost sampling location.

400 In 2014, TRA contributed about 18%, on average, to the simulated BC in snow, DOM
401 28%, FLR 44%, whereas ENE and IND were less significant. Maxima of TRA, DOM, and
402 FLR contributions were observed at a latitude of about 65°N, where measured EC and
403 modelled BC were similar. An example of the contribution from the aforementioned
404 dominant sources to snow BC concentrations for the highest measured EC concentration in
405 snow is shown in Figure 3. The transport sector includes emissions from all land-based
406 transport of goods, animals and persons. It is more significant in southern Russia and close to
407 the borders with Kazakhstan and Mongolia, where a large number of major Russian cities
408 (e.g., Moscow, Kazan, Samara, Yekaterinburg, Tomsk, Novosibirsk, Krasnoyarsk, etc...) are
409 located and connected with each other by federal highways. Residential and commercial
410 combustion includes emissions from combustion in households and public and commercial
411 buildings. Therefore, it is expected to be high for areas that consist of large population centres
412 (Figure 3). FLR emissions were found to contribute the most in this example with a total
413 concentration from this sector of 19.7 ng g⁻¹ (compared with 12.6 and 16.5 ng g⁻¹ in TRA and
414 DOM, respectively) (Figure 3).

415 In the Kindo Peninsula and in Arkhangelsk, where snow sampling took place in 2015,
416 the main contributions to snow BC were from DOM (47%), TRA (30%), BB (7%), and FLR
417 (6%) (see Figure 2). Similar to EC measurements in snow, simulated BC was also higher than
418 in 2014, as the sampling sites were located closer to strong sources in Europe (Kindo) and
419 close to a populated area (Arkhangelsk) with a strong regional impact. The highest
420 concentration of EC was observed in the Kindo Peninsula (33.13°E – 66.53°N). Figure 4
421 shows the spatial distribution of emissions that contributed to simulated snow BC at the
422 sampling point where the highest BC concentration was observed. In this case, TRA and
423 DOM emissions from Europe mostly affected snow in the Kindo Peninsula, whereas FLR
424 emissions were very low due to the long distance from the sampling point. Emissions from an
425 unusual late winter/early spring episode of BB in the borders of Belarus, Ukraine and Russia
426 also affected BC concentrations in snow in northwestern European Russia (Figure 4). The
427 importance of episodic BB releases in Russia, the miscalculation of satellite retrieved BB

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emissions and their impact in Arctic concentrations in early spring has been explained by Evangeliou et al. (2016) and Hao et al. (2016). BB emissions, originating mostly from Eastern Europe, contributed about 19.4 ng g^{-1} to the snow concentration at the receptor point (Figure 4). TRA and DOM emissions were the dominant sources for this sampling point, contributing 33.6 and 47.2 ng g^{-1} , respectively (Figure 4).

Finally, in 2016, when samples were collected at the Kindo Peninsula, in Arkhangelsk and in Yamal, DOM, FLR and TRA contributed, on average, 31% , 29% and 27% , respectively (see Figure 2(c)). Similar to the measured EC concentrations in snow, simulated concentrations of BC in 2016 were lower than those in 2015, on average. The highest measured EC concentration was observed in the Khanty-Mansiysk region ($72.94^{\circ}\text{E} - 65.36^{\circ}\text{N}$), which mirrors the simulated BC concentration at the same point very well. The much higher contribution from TRA at this sampling point (38.6 ng g^{-1}) (Figure 5(b)) is attributed to emissions from Southern Russia (e.g., Tomsk), where all the main cities in Russia are located. Another large fraction of TRA emissions comes from Central and Eastern Europe (see also in http://niflheim.nilu.no/NikolaosPY/SnowBC_141516.py). Similar to TRA, emissions from DOM were mostly transported to Khanty-Mansiysk from Central and Eastern Europe, as well as from Turkey contributing 36.6 ng g^{-1} (Figure 5). As previously mentioned, the sampling point where the highest EC concentration was measured is located inside the largest gas flaring region of Russia. In addition, the corresponding emission sensitivity maps showed that the air was coming from south passing directly through this high emission region making FLR emissions the highest contributing source (88.8 ng g^{-1}) (Figure 5).

4 Discussion

4.1 Cross validation of modelled BC concentrations with public datasets

In this section, we present an effort to further validate our model calculations of BC concentrations in snow. For this purpose, BC concentrations in snow that were adopted from Doherty et al. (2010) were compared with modelled BC concentrations in snow that were simulated with FLEXPART as described in section 2.4. Samples were collected in Alaska, Canada, Greenland, Svalbard, Norway, Russia, and the Arctic Ocean during 2005–2009, on tundra, glaciers, ice caps, sea ice, frozen lakes, and in boreal forests. Snow was collected mostly in spring, when the combination of snow cover and exposure to sunlight is at maximum and before the snow had started to melt. Samples of melting snow collected in the

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494 summer of 2008 from Greenland and from Tromsø, Norway, were removed from the study, as
495 we have no knowledge about the depth of the melt layer and effects of the percolation of
496 meltwater through the snowpack. All samples were collected away from local sources of
497 pollution. In many locations (Canadian Arctic, Russia, Greenland, Tromsø and Ny-Ålesund)
498 samples were gathered at different depths throughout the snowpack, giving information on the
499 seasonal evolution of BC concentrations as the snow accumulated (and/or sublimated)
500 throughout the winter. In these cases only the surface BC was taken into account. The snow
501 was melted and filtered, and the filters were analysed in a specially designed
502 spectrophotometer system to infer the concentration of BC (for more information see Doherty
503 et al., 2010). In contrast to our findings for the origin of snow BC in the Russian Arctic, a
504 source apportionment analysis perform in the 2008 and 2009 measurements (Hegg et al.,
505 2010) from this dataset showed that the dominant source of BC in the Arctic snow pack was
506 biomass burning. Specifically in Eastern Siberia biomass burning of crops and grasslands
507 contributed more snow BC in high latitudes than boreal forest fires, in contrast to the
508 Canadian Arctic.

509 A comparison of modelled (FLEXPART) and measured BC concentrations (Doherty et
510 al., 2010) in snow is depicted in Figure S 2. The model captures snow BC concentrations
511 relatively well in most of the Arctic regions except for the Canadian Arctic, where the
512 modelled concentrations of snow in 2007 were significantly higher. Samples from the same
513 region in other years showed moderate agreement with modelled values. Similar to our
514 finding for the new Russian measurements, the model output, with a MFB of -51%, tends to
515 underestimate deposition. The RMSE was estimated to be 52 ng g⁻¹, which is acceptable
516 considering that the variation of snow concentrations in the dataset ranged from 0.3 to 783 ng
517 g⁻¹. The highest measured concentrations of snow BC were observed in Russia, where the
518 model showed a good spatial agreement. For instance, the highest values were obtained in
519 Western Siberia, close to the gas flaring regions of the Nenets/Komi oblast, as well as in
520 southeastern and northeastern Russia, where air masses were arriving from high emitting
521 sources in southeastern Asia. Lower biases in modelled BC concentrations were observed in
522 northern Siberia with the exception of a few samples at the coasts of the Kara Sea and
523 northeastern Siberia. Furthermore, biased BC concentrations were also observed in Greenland
524 and northern Canada. In Western Siberia, BC in snow presented in Doherty et al. (2010)
525 between 2005–2009 was 101±153 ng g⁻¹ on average, which is very close to the average value
526 of measured EC obtained from the sampling 2014–2016 campaigns (83±37 ng g⁻¹).

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536 From total number of samples presented in (Doherty et al., 2010) that were used here
537 for validation, only six were collected in the Yamal Peninsula similar as part of the data
538 presented in the current paper. The rest was collected in Nenets/Komi region and in Eastern
539 Russia and cannot be directly compared with snow EC measurements from the 2014 – 2016
540 campaigns. BC concentrations in Yamal Peninsula in 2007 ranged from 4.1 to 17.6 ng g⁻¹
541 (Average±SD: 10.1±4.8 ng g⁻¹). In the same region, we report EC concentrations to be more
542 than double varying between 6.6 to 55 ng g⁻¹ (Average±SD: 25.7±15.8 ng g⁻¹), whereas there
543 were two samples that showed EC concentrations of more than 100 ng g⁻¹. As mentioned in
544 section 2.1 the sampling of snow for the EC analysis took place more than 500 m away from
545 roads to minimize influence from traffic emissions, while a similar statement is also found in
546 the (Doherty et al., 2010) data. Nevertheless, considering that the samples were not collected
547 from the same regions exactly and at the same time, no safe conclusions can be obtained.

548 Modelled BC concentrations simulated with FLEXPART were also compared with
549 snow BC concentrations from samples collected at the Global Atmosphere Watch
550 Observatory at Alert, Nunavut, from September 14th, 2014 to June 1st, 2015 and they are
551 available in Macdonald et al. (2016). Alert is a remote outpost in the Canadian high Arctic, at
552 the northern coast of Ellesmere Island (82°27' N, 62°30' W), with a small transient
553 population of research and military personnel. Sampling details and analytical methodologies
554 used for the analysis of BC can be found in Macdonald et al. (2016). BC concentrations in
555 FLEXPART were simulated as in all previous analyses described in this paper (see section
556 2.4.). Timeseries of simulated and measured BC are depicted in Figure S 3, for the whole
557 sampling period. As before, a correlation coefficient (R) of 0.63 indicates that our model
558 captures the temporal variation of the measured BC in snow. The RMSE was estimated to be
559 almost 63 ng g⁻¹, a relatively high value. The MFB of 47% indicates a strong overestimation
560 of snow concentrations, although in many samples the opposite was also observed (Figure S
561 3). This is in contrast to the previous data sets discussed, for which the model underestimated
562 measurements.

563 Further analysis was carried out to adequately understand the origin of the
564 aforementioned overestimations in the Canadian Arctic in both datasets (Doherty et al., 2010;
565 Macdonald et al., 2017), as they are shown to be rather systematic. For this reason, we have
566 calculated the average footprint emission sensitivities and the average BC contribution from
567 the major sources in ECLIPSE for the 2007 snow samples in the Canada Arctic and for Alert
568 samples. We have chosen these samples, because they were three or more times higher than

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586 the observations, and in this way we can, locate the observed overestimations predicted with
587 FLEXPART (Figure 6).

588 Regarding the model overestimation for the 2007 samples, the average footprint
589 emission sensitivity showed that the air was coming from continental regions of Canada with
590 a smaller contribution from Scandinavia (Figure 6). The highest emission sources for these
591 samples were TRA and DOM that contributed almost 80% to the snow concentrations,
592 whereas forest fires were less important at the time of sampling. Two hot spots were
593 identified, one along the borders of Canada with USA and another, of smaller intensity, in
594 southeastern Asia. A similar emission sensitivity was obtained for the same area of the
595 Canadian Arctic in 2009 only slightly shifted to the north, simulated concentrations were in
596 very good agreement with observations (Figure S 2). This shows that the model
597 overestimation for the 2007 samples is likely attributed to an overestimation of TRA and
598 DOM sources in North America in ECLIPSE for 2007. For the Alert samples, for which the
599 model strongly overestimated BC, the major sources were TRA and FLR, which contributed
600 55%, and BB which contributed about 7 ng g⁻¹ (22%) on average (Figure 6). Anthropogenic
601 BC arriving from Europe and Russia has been previously shown to be important for Alert air
602 pollutant concentrations (Sharma et al., 2013). The model overestimation of BC in snow
603 samples at Alert needs further investigation. It is likely that it originates from anthropogenic
604 emissions in northwestern America or in Europe, because forest fires in Canada and Russia,
605 although important for Alert (e.g., Qi et al., 2017), were not significant in the present
606 comparison.

607 4.2 Model deviation from snow EC measurements and region-specific 608 contribution of sources

609 It has been shown that, measured concentrations of EC in snow in northwestern
610 European Russia and Western Siberia were underestimated in FLEXPART (Figure 2). This
611 was confirmed by the calculated fractional bias (see section 3.2), the spatial distribution of
612 which is shown in Figure S 1. To examine whether this underestimation was due to missing
613 emission sources or errors in modelled transport and deposition, we have calculated the
614 average footprint emission sensitivity for those sampling points, for which FLEXPART
615 strongly ($FB < -100\%$) and slightly ($-100\% < FB < 0\%$) underestimated the observed
616 values. The average footprint emission sensitivities are shown in Figure 7, together with the
617 locations of active fires in the last two months before the sample collection. The fire data

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641 | were adopted from MODIS (Moderate Resolution Imaging Spectroradiometer) (Giglio et al.,
642 | 2003) and the gas flaring facilities from the Global Gas Flaring Reduction Partnership
643 | (GGFR) (<http://www.worldbank.org/en/programs/gasflaringreduction>).

644 | When the model strongly underestimated the measured EC ($FB < -100\%$), the
645 | average footprint emission sensitivity showed the highest values over the Yamal Peninsula
646 | and the agglomeration of many gas flares in Khanty-Mansiysk (Figure 7, (b)). This might
647 | confirm the finding of Huang et al. (2014) that gas flaring emissions in the ECLIPSE
648 | inventory, while very high, are still underestimated. According to a related study by Huang
649 | and Fu (2016), Russia contributes 57% to the global BC emissions from gas flaring,
650 | Underestimation of modelled atmospheric concentrations compared to observations from the
651 | Barents and Kara Seas was recently also reported by Popovicheva et al. (2017), although the
652 | underestimation was relatively small.

653 | When FLEXPART showed a moderate underestimation of EC concentrations in snow
654 | ($-100\% < FB < 0\%$), the emission sensitivity was high near Arkhangelsk and over
655 | Scandinavia (Figure 7). BC emissions in Scandinavia are considered relatively low in most
656 | inventories and contribute no more than 6.5% to the global emissions in ACCMIP (Aerosol
657 | Chemistry Climate Model Intercomparison Project) (Lamarque et al., 2013), 6.2% in
658 | EDGARv4.2 (Emission Database for Global Atmospheric Research) (Olivier et al., 2005),
659 | 2.1% in MACCity (Monitoring Atmospheric Composition & Climate / megaCITY - Zoom for
660 | the ENvironment) (Hollingsworth et al., 2008; Stein et al., 2012) and 3.3% in ECLIPSE
661 | (Klimont et al., 2016). The highest emission sensitivity was found over northwestern Russia
662 | (Figure 7), a region which includes Murmansk, Pollution levels in Murmansk could be high
663 | due to emissions from local industry, mining, heating and transport (Law and Stohl, 2007).
664 | Another potential source region was Nenets/Komi area and Western Kazakhstan, where a few
665 | other flaring facilities are located (Figure 7).

666 | Figure 7 shows that the underestimation of observed EC concentrations in snow
667 | strongly depends on the region, where samples are collected. In Western Siberia, the
668 | underestimation was larger than in northwestern European Russia. For this reason, we have
669 | computed the average region-specific emission sensitivities and the average region-specific
670 | contribution from the major polluting sources identified in ECLIPSE dataset. We distinguish
671 | between three regions, northwestern European Russia, Western Siberia (north of 62 °N) and
672 | Western Siberia (south of 62 °N) (Figure S 4– S 6). For the samples collected in northwestern

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687 European Russia (Figure S 4), an average contribution of 21.6 ng g⁻¹ from all sources was
688 estimated to have originated mainly from TRA (7.7 ng g⁻¹) and DOM (10.4 ng g⁻¹) sources in
689 Finland. The contribution from BB and FLR emissions was insignificant (8% and 6%,
690 respectively), whereas the rest of the ECLIPSE sources were negligible (IND, ENE, WST).
691 For the samples collected at high latitudes in Western Siberia, the average contribution from
692 all sources was more than 4 times higher (86 ng g⁻¹) than those observed in northwestern
693 European Russia (Figure S 5). FLR emissions accounted for 40% of the total contribution,
694 which reflect the proximity of the sampling site to the main flaring facilities of Russia. The
695 average contribution from TRA activities in Europe and southeastern Russia to the northern
696 part of Western Siberia was 24%. Finally, DOM emissions in Eastern Europe also contributed
697 another 28%. Finally, for the samples that were collected in the southern part of the Western
698 Siberia, an average contribution of 47.4 ng g⁻¹ was estimated from all sources included in
699 ECLIPSE (Figure S 6). The highest contributing categories were TRA and DOM, whereas
700 FLR appeared to contribute less, although the sampling site is close to Khanty-Mansiysk
701 flaring region. This is attributed to the prevailing winds that forced flaring emissions to a
702 northernmost direction opposite to the location of the sampling stations (see Figure S 6).

703 Overall, the region-specific analysis of the sources contributing to modelled BC in
704 snow showed that the DOM, FLR and/or TRA sources might explain the model
705 underestimation in high Arctic. However, in the most recent assessments of BC of the higher
706 Arctic (Popovicheva et al., 2017; Winiger et al., 2017), it was shown that ECLIPSE captures
707 levels of BC quite well, whereas FLR emissions might have a smaller impact in the Central
708 Siberian Arctic (Tiksi) than previously estimated. Surprisingly, the average contribution from
709 BB in lower latitudes was extremely low in all Western Siberia (Figure S 5, and S 6), despite
710 the fact that sampling took place in springtime, where BB becomes important. Evangeliou et
711 al. (2016) reported that using a different dataset, that is based on the same approach as GFED,
712 but includes updated emission factors for Eurasia, surface concentrations of BC in the Arctic
713 stations can be substantially higher. This shows the need for further investigation of BC
714 sources in the Russian Arctic.

715 5 Conclusions

716 We have analysed snow samples collected in Western Siberia and northwestern
717 European Russia in 2014, 2015 and 2016 with respect to EC. This region is of major interest
718 due to its large uncertainty in BC emissions and because it is located in the main transport

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736 route of BC to the Arctic. An effort to constrain the sources that contribute to measured
737 concentration in BC in snow was made using the LPDM FLEXPART (version 10).

738 The observed EC levels in snow varied widely within and between regions (3–219 ng g⁻¹
739 ¹ for 2014, 46–175 ng g⁻¹ in 2015 and 7–161 ng g⁻¹ in 2016), and are in the upper range of
740 previously reported concentrations of EC and BC in snow in the Arctic region. However, the
741 observed levels presented here appear typical for Western Siberia, which is subject to high
742 domestic Russian emissions as well as to transport from distant European ones.

743 The snow BC concentrations predicted by the model are in a fair agreement with EC
744 observations over Western Siberia and northwestern European Russia ($R = 0.5 - 0.8$).
745 However, the calculated MFB values (-48% to -27%) showed that the model systematically
746 underestimated observations in Russia. This underestimation strongly depended on the region
747 where the samples were collected. In northwestern European Russia, the main contributing
748 sources were TRA and DOM mainly from adjacent regions in Finland. TRA and DOM
749 contributed double to snow BC sampled at low latitudes of Western Siberia (<60°N) as
750 compared to samples collected over regions above 60°N; the majority of these emissions
751 originating from highly populated centres in Central Europe. Finally, in higher latitudes of
752 Western Siberia (>60°N), snow BC concentrations were further increased mainly due to FLR
753 emissions from facilities located close to the snow sampling points.

754 The modelled BC concentrations in snow were further investigated using two
755 independent public measurement datasets that include samples from all over the Arctic for the
756 period 2005 to 2009 and from Alert in 2014 and 2015. The model captured levels of BC fairly
757 well despite the large variation in measured concentrations. An exception was observed in
758 North America in spring 2007 and in Alert observatory in late winter – early spring 2015. In
759 both cases, the major sources were along the Canadian borders with USA and in Western
760 Europe. Considering the fact that similar deviations were not observed in samples collected in
761 the area during other years, it is likely that some of the prevailing sources of BC in this region
762 show strong temporal variability in their emissions, and this is not taken into account in
763 ECLIPSE inventory. Previously reported average measurements of BC concentrations in
764 snow in Western Siberia and northwestern European Russia were $101 \pm 153 \text{ ng g}^{-1}$, which is
765 about 20% higher than the EC measurements presented here ($83 \pm 37 \text{ ng g}^{-1}$).

766 *Data availability.* All data used for the present publication can be obtained from the
767 corresponding author upon request.

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781 *Competing interests.* The authors declare that they have no conflict of interest.

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795 interactive website for fast visualization
796 (http://niflheim.nilu.no/NikolaosPY/SnowBC_141516.py). All results can be accessed upon
797 request to the corresponding author of this manuscript.

798 *Author Contributions.* N. Evangeliou designed and performed the modelling experiments
799 and wrote the paper. V. P. Shevchenko organised and performed the sampling of EC, K.-E.
800 Yttri performed all the TOA of the snow samples. S. Eckhardt modified FLEXPART model
801 for the calculation of footprint emission sensitivities for deposited mass. E. Sollum wrote an
802 algorithm that computes the starting date of the FLEXPART releases based on the water
803 equivalent volume from ECMWF. O. S. Pokrovsky, V. O. Kobelev, V. B. Korobov, A. A.
804 Lobanov, D. P. Starodymova and S. N. Vorobiev assisted the sampling campaigns in Western
805 Siberia and northwestern European Russia during 2014–2016. R. L. Thompson and A. Stohl
806 supervised the study and wrote parts of the paper.

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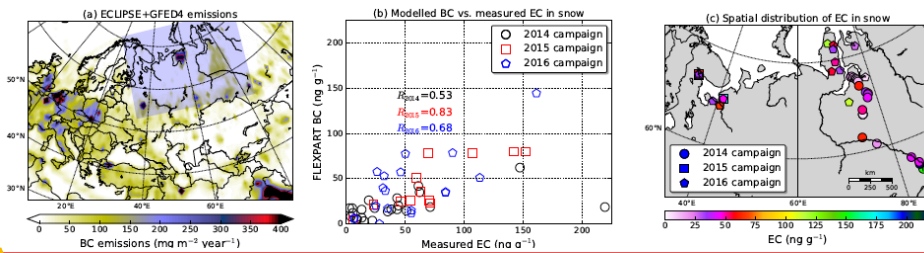
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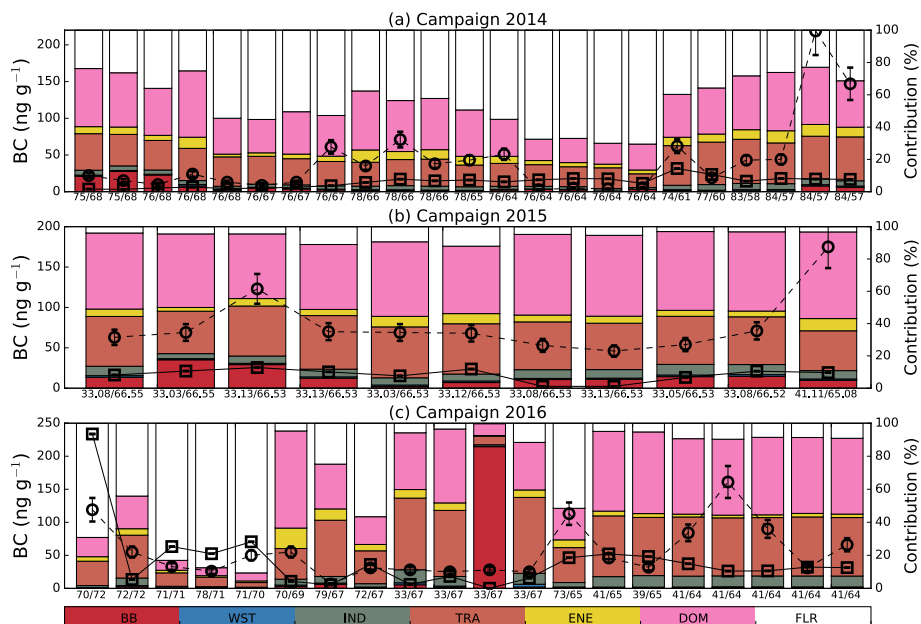
1023 **Figure 1.** (a) Total emissions of BC (anthropogenic emissions from ECLIPSE (Klimont et al.,
 1024 2016) and biomass burning from GFED4 (Giglio et al., 2013). The blue shade shows the area
 1025 of interest that is zoomed on the right. (b) Comparison of modelled BC concentrations in
 1026 snow with measured EC concentrations. (c) Spatial distribution of EC in snow measured by
 1027 thermal optical analysis (TOA) of filtered snow samples from northwestern European Russia
 1028 and Western Siberia in spring-time 2014, 2015 and 2016.

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SOURCE CONTRIBUTION TO SNOW BC



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1037 **Figure 2.** Contribution from the various emission categories considered in the ECLIPSE and
 1038 GFED inventories to simulated BC concentrations in snow in (a) 2014, (b) 2015 and (c) 2016
 1039 in Western Siberia and northwestern European Russia. BB stands for biomass burning, WST
 1040 for waste burning, IND for industrial combustion and processing, TRA for surface
 1041 transportation, ENE for emissions from energy conversion, and extraction, DOM for
 1042 residential and commercial combustion, and FLR for gas flaring. Bars show the relative
 1043 source contribution (0 –100%, right axis) and are sorted, from left to right, from the
 1044 northernmost to the southernmost measurement location (coordinates are reported on the
 1045 bottom as longitude/latitude). Measured EC concentrations in snow are reported with open
 1046 circles, whereas modelled BC is shown with open rectangles (left axis).

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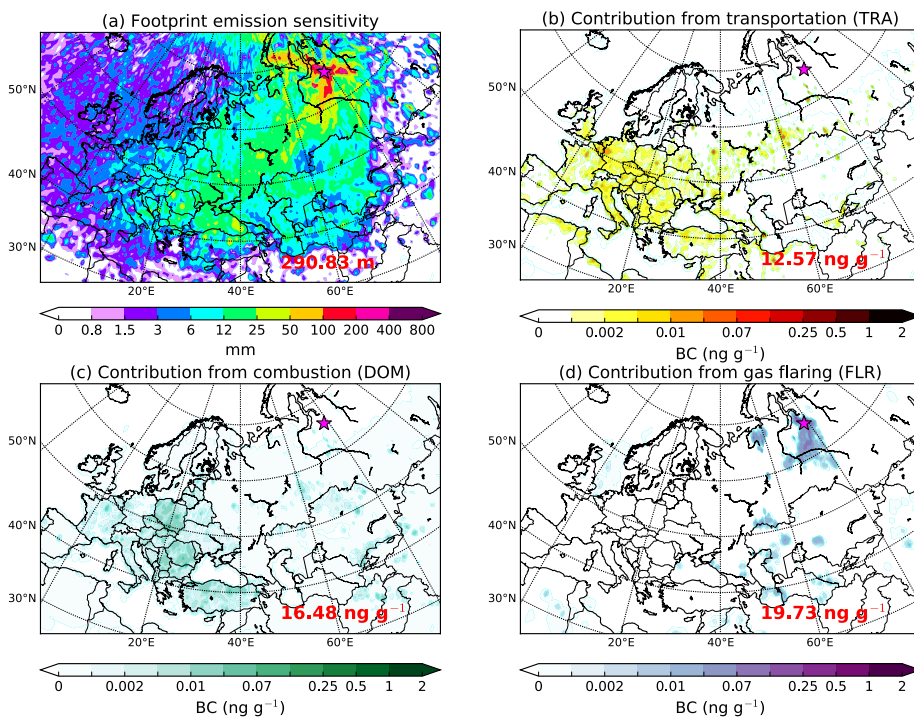
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**EMISSION SENSITIVITY AND SOURCE CONTRIBUTION TO SNOW BC IN 2014
(78.17° E - 65.78° N)**



1055

1056 **Figure 3.** (a) FLEXPART emission sensitivity, contribution from (b) transportation (TRA),
 1057 (c) residential and commercial combustion (DOM) and (d) gas flaring (FLR) to the maximum
 1058 measured concentration of snow EC recorded along the transect from Tomsk to Yamal
 1059 Peninsula in Western Siberia during the campaign of 2014.

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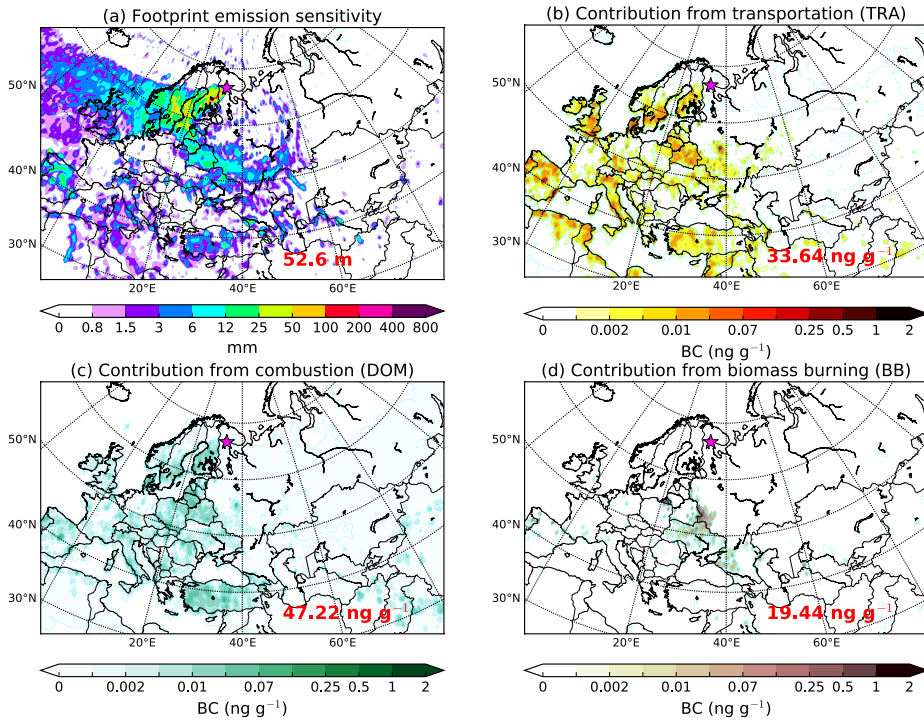
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**EMISSION SENSITIVITY AND SOURCE CONTRIBUTION TO SNOW BC IN 2015
(33.13° E - 66.53° N)**



1065

1066 **Figure 4.** (a) FLEXPART emission sensitivity, (b) contribution from transportation (TRA),
 1067 (c) residential and commercial combustion (DOM) and (d) gas flaring (FLR) to the maximum
 1068 measured concentration of snow EC recorded in northwestern European Russia (Kindo
 1069 Peninsula and Arkhangelsk region) during the campaign of 2015.

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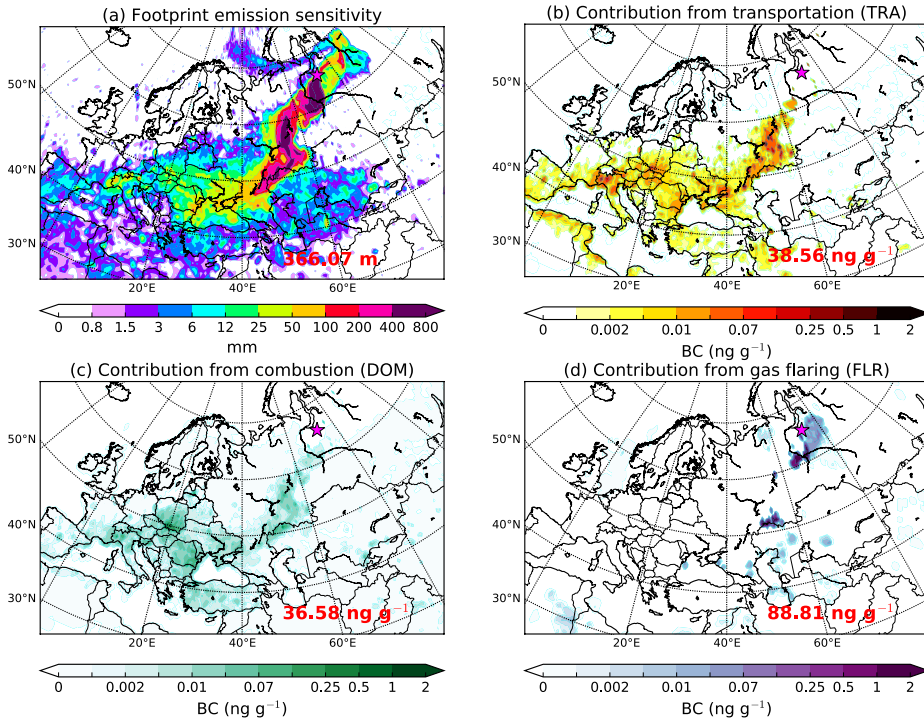
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**EMISSION SENSITIVITY AND SOURCE CONTRIBUTION TO SNOW BC IN 2016
(72.94° E - 65.36° N)**



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1076 **Figure 5.** (a) FLEXPART emission sensitivity and (b) contribution from transportation
 1077 (TRA), (c) residential and commercial combustion (DOM) and (d) gas flaring (FLR) to the
 1078 maximum measured concentration of snow EC recorded in Kindo Peninsula, Arkhangelsk and
 1079 Yamal Peninsula (northwestern European Russia, Western Siberia) during the campaign of
 1080 2016.

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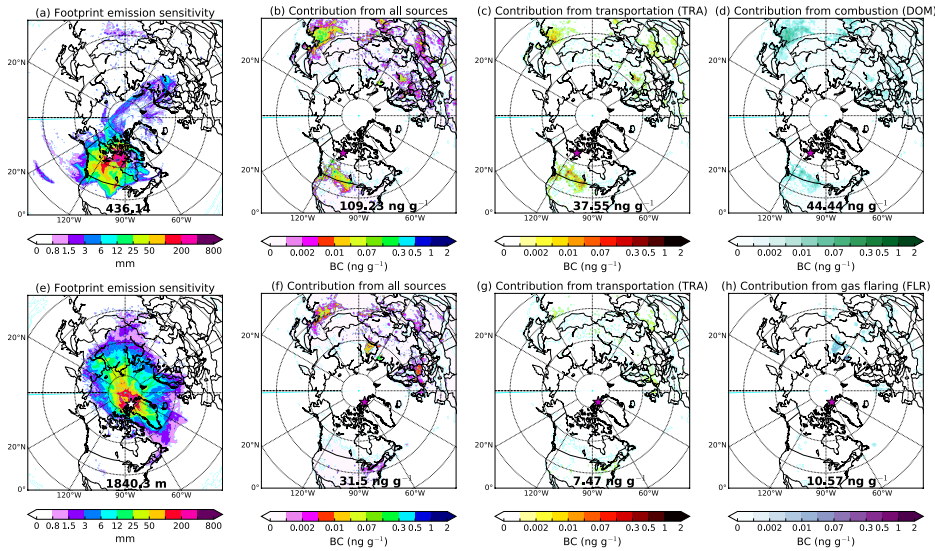
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**EMISSION SENSITIVITY AND SOURCE CONTRIBUTION TO SNOW BC
(CANADIAN ARCTIC 2007 - ALERT 2014-2015)**



1086

1087 **Figure 6.** (a–d) Footprint emission sensitivity and major contribution from all sources, TRA
 1088 and DOM averaged for the samples that showed overestimated modelled concentrations of
 1089 BC in 2007 (Doherty et al., 2010). (e–h) Footprint emission sensitivity and contribution from
 1090 all sources, TRA and FLR for the samples collected in Alert (Macdonald et al., 2017) that
 1091 model overestimated by more than three times.

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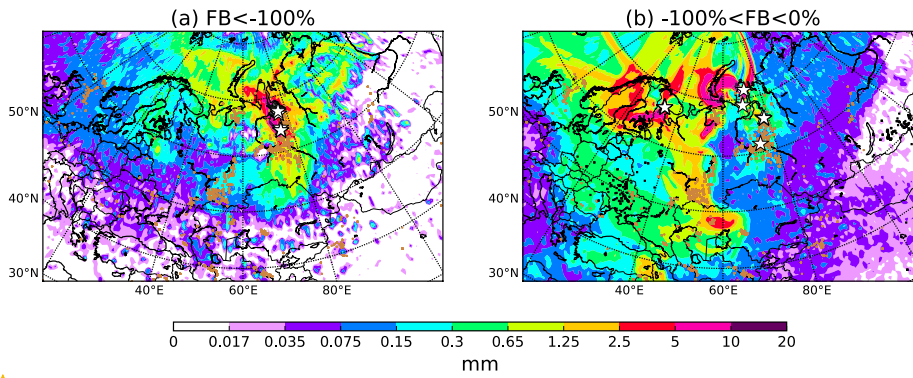
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**AVERAGE FOOTPRINT EMISSION SENSITIVITY
NORMALISED AGAINST UNDERESTIMATION FROM OBSERVATIONS**



1095

1096 **Figure 7.** (a) Footprint emission sensitivity from FLEXPART averaged for the sampling
1097 points where the model underestimated observations significantly ($FB < -100\%$) and (b)
1098 less significantly ($-100\% < FB < 0\%$). Black squares show the locations of active fires
1099 detected by MODIS (Moderate Resolution Imaging Spectroradiometer) (Giglio et al., 2003).
1100 Brown dots show the location of gas flaring sites from the Global Gas Flaring Reduction
1101 Partnership (GGFR) (<http://www.worldbank.org/en/programs/gasflaringreduction>).

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1103 **FIGURE CAPTIONS FOR SUPPLEMENTS**

1104

1105 **Figure S 1.** Fractional bias ($FB = [(C_m - C_o)/(C_m + C_o) \times 0.5] \times 100\%$) for all samples
1106 collected from the three campaigns in Western Siberia and northwestern European Russia in
1107 2014, 2015 and 2016. MFB (mean fractional bias) is the fractional bias averaged for all snow
1108 samples from 2014, 2015 and 2016, whereas RMSE is the root mean square error in $ng\ g^{-1}$.

1109 **Figure S 2.** (a) Distribution of snow measurements of BC adopted from Doherty et al. (2010)
1110 in the Arctic from 2005 to 2009, (b) Simulated (FLEXPART) BC concentrations in snow for
1111 the same period (right). MFB, RMSE and correlation coefficient (R) values are further given.

1112 **Figure S 3.** Timeseries of simulated and measured BC concentrations in snow collected in
1113 Alert (Macdonald et al., 2017). Correlation coefficient (R) between modelled and measured
1114 BC, RMSE and MFB values are also shown.

1115 **Figure S 4.** (a) Average footprint emission sensitivity and (b-f) source contribution (from all
1116 sources, TRA, DOM, FLR and BB) for all the samples located in northwestern European
1117 Russia.

1118 **Figure S 5.** (a) Average footprint emission sensitivity and (b-f) source contribution (from all
1119 sources, TRA, DOM, FLR and BB) for all the samples located in Western Siberia (north of 62
1120 °N).

1121 **Figure S 6.** (a) Average footprint emission sensitivity and (b-f) source contribution (from all
1122 sources, TRA, DOM, FLR and BB) for all the samples located in Western Siberia (south of
1123 62 °N).

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