

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

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General Comments 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. In-

C1

deed 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.

C2

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.

C3

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 as 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

C4

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”

C5

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

C6

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 ultra-pure 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.

C7

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.

C8

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

C9

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 concentrations"

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

C10

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.

C11

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, choice 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?

C12

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.

C13

Response: Corrected.

70. Line 374-375: should read” Similar to our finding for the new Russian measurements, 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.

C14

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

C15

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-Mansijsk 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/Mansijsk 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 similar 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)

C16

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.

C17

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 similar"

C18

.observed 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.

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

C19

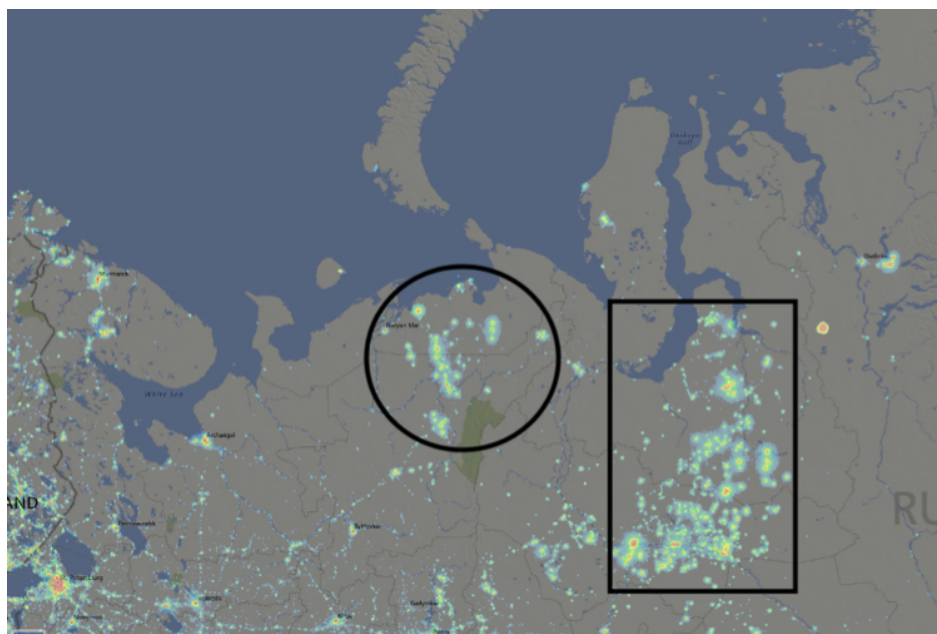


Fig. 1.

C20