

Interactive comment on “Top–down estimates of black carbon emissions at high latitudes using an atmospheric transport model and a Bayesian inversion framework” by Nikolaos Evangeliou et al.

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This work is an attempt to constrain the BC emissions at high northern latitudes starting from four different inventories and applying a Bayesian inversion to the model Flexpart. The choices of emission inventories, of the surface stations and of ancillary BC data to independently optimize BC emissions are well explained. Additional information is gained through varying below-cloud and in-cloud scavenging to adapt BC-like tracers that are used to improve the a-posteriori BC concentrations simulated by Flexpart. This paper is well structured, the approach is interesting and the results are pertinent.

C1

I suggest that it can be published after several improvements to the wording that are outlined below. I also have a few suggestions for reinforcing the discussion.

RESPONSE: We would like to acknowledge reviewer for his positive comments on our paper and his kind willingness to help improving this manuscript. We have tried to follow all of his suggestions.

Lines 314-315 page 11: “The different scavenging coefficients used did not create a large variation in the monthly concentrations of BC. The best performance for the majority of the stations examined and most months was obtained for species 1, 2 and 10 (see Table 2).” This is a rather surprising result since wet scavenging is the main global sink for BC. Does that indicate that dry deposition is an important process for removing BC at high northern latitude, in that case why didn't the authors vary the efficiency of dry scavenging as well as wet scavenging? This aspect deserves several sentences in order to develop why the authors obtained such quantitative result.

RESPONSE: The reviewer has a good point in his comment. However, still dry deposition is only 20% of the total as in most global models.

The small variability stems from the small perturbation of the wet scavenging parameters. When changing scavenging parameters of BC, except for conducting a sensitivity analysis and estimate uncertainty, our top priority was to select the best representative species for BC, hence scavenging coefficients should be realistic. This resulted in the aforementioned small variability. Of course, if we change these parameters drastically, the variability will increase, but then we do not have realistic values anymore. We have now tried to clarify this in the text (please check manuscript with Track Changes).

Figure 7 shows that vertical profiles can be optimized by the Bayesian approach. Schwartz et al. (2013) illustrate well that modelled BC concentrations profiles are systematically overestimated in the mid- to high-troposphere over the remote Pacific. The biases are much less pronounced over source regions. What can be learned from this work about improving the model performances over remote regions?

C2

RESPONSE: The Bayesian approach forces emissions mathematically, in order to better match observations. Like we explain in section 3.4, the comparison to observations included in the inversion is not a sufficient indicator of the inversion's performance, as the inversion is designed to reduce the model-observation mismatches. The magnitude of the posterior reduction of the model mismatch to the observations is partly determined by the weighting given to the observations relative to the prior emissions. A much better performance indicator is the comparison of the posterior concentrations with observations that were not included in the inversion (independent observations). For this reason, we have chosen to compare our posterior emissions with profiles from the ACCACIA campaign, which were not included in the inversion, and they were available in public for year 2013. We did exactly the same for years 2014 and 2015. So, the use of these profiles was for validation only.

Now, likewise in Schwartz et al. (2013) paper, our model overestimates up to 3 times concentrations of BC at lower altitudes, while this overestimation becomes more intense at higher altitudes. What we claim in the paper is that the observed model overestimation decreases drastically when using the resulting a posteriori emissions that were calculated from our Bayesian inversion framework (based on ground-based measurements).

In the abstract, I recommend that the authors substitute "posterior emissions" with "a-posteriori emission estimates" since at this stage the reader does not know what "posterior emissions" refers to.

RESPONSE: We very well agree and have corrected the manuscript (see manuscript with Track Changes).

Reference: Schwarz JP, Samset BH, Perring AE, et al. Global-scale seasonally resolved black carbon vertical profiles over the Pacific. *Geophysical Research Letters*. 2013;40(20):5542-5547. doi:10.1002/2013GL057775.

Minor comments:

C3

Page 10, line 265: NSD was never defined before, I assume it refers to 'Normalized Standard Deviation' but the reader should not have to guess.

RESPONSE: We appreciate for this comment. We also noticed that nRMSE is not defined in the same paragraph. We have now corrected both (see manuscript with Track Changes).

Page 12, lines 334 & 337: the stations BOS (Bösel) and WLD (Whaldof) are discussed but not shown in Fig. S3

RESPONSE: As shown in Table 1, 17, 13 and 24 stations with continuous measurements were used for 2013, 2014 and 2015, respectively. It is not possible to show the results for all of them. Therefore we decided to show the northernmost stations. However, if we only comment on what we show, we are only telling half of the truth. Specifically, commenting on what it is shown only would result on missing a very important statement that all prior emission datasets failed to reproduce the observations in central Europe during all years studied (2013, 2014 and 2015) implying either missing sources or highly uncertain measurements (end of second paragraph of section 3.2).

Page 12, line 343: change "by 23% at maximum" to "by a maximum of 23%"

RESPONSE: Corrected (see manuscript with Track Changes).

Page 12, lines 344-353: change: "NMSE values calculated for each of the four emission inventories were very low at the majority of the stations for which data existed in all the years of study (ZEP, SUM, TIK, BAR, MEL and LEI), when ECLIPSEv5 emissions were used, while at PAL all emission datasets performed well (Figure 5). At most of the Arctic stations, the simulations using ECLIPSEv5 reproduced the observations better compared to the other inventories examined. This shows that the most appropriate emission dataset for our purpose is the ECLIPSEv5 inventory, as it is the only one that can capture the characteristically elevated concentrations of BC in the Arctic, which persist until spring, and are caused by anthropogenic emissions (Law and Stohl,

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2007). A significant deficiency is found for TIK for reasons that were explained earlier (see section 3.1).” to “Normalized mean square error values calculated for each of the four emission inventories were very low at the majority of the stations for which data existed in all the years of study (ZEP, SUM, TIK, BAR, MEL and LEI), when ECLIPSEv5 emissions were used. In contrast, at PAL all emission datasets performed well (Figure 5). The observations of BC concentrations at Arctic stations were better reproduced in simulations using the ECLIPSEv5 than with any other inventories examined. Law and Stohl (2007) have documented that these elevated BC concentrations are caused by anthropogenic emissions. Black carbon concentrations at TIK are not well simulated for reasons given in section 3.1.”

RESPONSE: Corrected according to reviewer’s suggestion (see manuscript with Track Changes).

Page 13, lines 366-367: change “Table 3 reports annual prior and posterior emissions of BC for different regions and 367 average emissions for the period 2013–2015.” To “Table 3 reports annual prior, posterior, and averaged over 2013-2015, BC emissions for different regions.”

RESPONSE: We have also corrected this sentence (please see manuscript with Track Changes).

Page 13, line 371 and everywhere in the remaining text: change “emissions of BC” to “BC emissions”.

RESPONSE: We have corrected the expression from page 13 onwards. To keep a level of diversity in the manuscript, we have kept the expression as it was before (“emissions of BC”) in the titles of sections and in the legends of figures (please see manuscript with Track Changes).

Page 14, line 410-411: change “The relative uncertainty of the inversion averaged over the period 2013 to 2015 was estimated to be 30%.” To “Averaged over the period

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2013-2015, the relative uncertainty of the inversion was estimated to be 30%.”

RESPONSE: Corrected according to reviewer’s suggestion (see manuscript with Track Changes).

Page 15, lines 458-460: change “In the same figures the differences between posterior and prior emissions (ECLIPSEv5) are shown (right panels) to indicate the biggest emission changes compared to the a priori dataset.” to “The right panel of the same figures shows the differences between posterior and prior emissions (ECLIPSEv5) and highlights the biggest emission changes compared to the a priori dataset.”

RESPONSE: Corrected according to reviewer’s suggestion (see manuscript with Track Changes).

Page 16, line 464: change “... was seen in 60°N- 135°W” to “was located at 60°N, 135° W”.

RESPONSE: Corrected (see manuscript with Track Changes).

Page 16, lines 474-476: change “In this region four uranium mines are located that use diesel generators, diesel trucks, and likely also other diesel-powered machinery.” to “Uranium mines are located in this region. These mines use diesel generators, diesel trucks, and other diesel-powered machinery.”

RESPONSE: Corrected according to reviewer’s suggestion (see manuscript with Track Changes).

Page 18, lines 544-545: change “By separating the inversion domain into continental regions, it is easily seen where biomass burning is important.” to “Separating the inversion domain into continental regions reveals where biomass burning is important.”

RESPONSE: Corrected according to reviewer’s suggestion (see manuscript with Track Changes).

Page 18, line 550: change “. . . largest peak was already in April. . .” to “. . .larges

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peak appears in April. . .”

RESPONSE: Corrected (see manuscript with Track Changes).

Page 19, line 563-565: change “We performed a sensitivity study to assess the best representative species for BC in terms of scavenging and removal and the best representative emission inventory to be used as the prior information for our inversion.” to “We performed a sensitivity study to assess the best representative species for BC according to the efficiency of in-cloud and below-cloud scavenging, and the best representative emission inventory to be used as the prior information for our inversion.”

RESPONSE: Corrected according to reviewer’s suggestion (see manuscript with Track Changes).

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