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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 by Evangeliou et al. performed a Bayesian inverse analysis of BC emission fluxes at northern high latitudes using ground-based observations and a Lagrangian particle model. The authors accounted for the uncertainties associated with BC wet removal with simulations using a range of scavenging parameters and prior emissions. The inversion found high BC emissions from gas flaring in Russia and Canada. The inversion also found that retrieved emission seasonality is different from that in emission inventory in N. America, N. Europe and N.Siberia. The topic is interesting and the paper is overall well organized but the method description still needs some clarification. I support the publication of the paper if the following major comments are addressed.

RESPONSE: We would like to acknowledge reviewer's attempt to help improving this paper. We have tried to correct and answer his comments.

–Major comments

–The paper lacks sufficient description about the specification of error covariance matrices, B and R, which are useful for readers to evaluate the validity of the inversion method.

RESPONSE: The diagonal elements of B (the variances) are the squares of the uncertainties of each grid cell where the uncertainties are calculated as a fraction of the total prior emission estimate with a lower limit. For R, normally this is a diagonal matrix (it only has off-diagonal elements if one uses the calculation of aggregation error). The diagonal elements of R are calculated as the quadratic sum of the observation, aggregation and background uncertainties. In our case we had zero for the background uncertainty (zero background for BC).

Details and specifications of the error covariance matrices can be found in Thompson and Stohl (2014). Prior to publication of this manuscript in ACPD, we were asked many times to change the methodology of the manuscript, in order to pass the similarity test. Therefore, we were encouraged to omit many methodological details that can be found in the aforementioned paper.

Thompson, R. L. and Stohl, A.: FLEXINVERT: an atmospheric Bayesian inversion framework for determining surface fluxes of trace species using an optimized grid, Geosci. Model Dev., 7, 2223-2242, https://doi.org/10.5194/gmd-7-2223-2014, 2014.

–How large is the observation errors and prior errors? Are they assumed to have any spatial/temporal correlations? Are model errors considered in R?

RESPONSE: As described in Thompson and Stohl (2014) paper the inversion code treats observation error in two ways. Either one can assign a constant observation error or take observation error directly from the continuous measurements. We have chosen the second option, because we think that observation errors that have been given by the instruments are far more realistic.

As described in the paper released for the ECLIPSE emissions by Klimont et al. (2017), there are different types of uncertainties in ECLIPSE that stem from the way that each calculation has been made. For example for residential combustion, there are uncertainties in assumptions about heat value of various biofuels. For emissions from waste burning large uncertainties are attributed to only scarce measurements and difficulties in finding reliable data on waste collection, recycling, and disposal rates. Large uncertainties exist in fuel consumption, its allocation between uses and technologies, and emission factors. Several other sources of uncertainty are also mentioned in Klimont et al. (2017) paper. This makes impossible to estimate a fixed uncertainty of the prior. We have used a minimum and maximum error. Then the prior error is calculated as a fraction of the maximum value of each pixel and its 8 surrounding ones, and then sets ocean pixels to the minimum value (see previous comment and info in Thompson and Stohl, 2014).

Error covariance matrix B has spatial and temporal correlations (off-diagonal elements). About the model errors, these are represented by the ensemble of runs that we have performed. Each member of the ensemble has different scavenging settings, so the range of results from our ensemble represents the range due to the uncertainty in the scavenging settings, which in our case is the largest model error.

Klimont, Z., Kupiainen, K., Heyes, C., Purohit, P., Cofala, J., Rafaj, P., Borken-Kleefeld, J., and Schöpp, W.: Global anthropogenic emissions of particulate matter including black carbon, Atmos. Chem. Phys., 17, 8681-8723, https://doi.org/10.5194/acp-17-8681-2017, 2017.

–Also, I find the method description a little confusion. In Line 191, the authors claim that they conduct "ensemble of inversions" and conduct "the inversion for BC represented by 12 different scavenging coefficients and for four different prior emission datasets. However, in Line 358, "the posterior emissions were calculated for the best performing species and best prior emission inventory". So, what are the results of "ensemble of inversions" used for? Are they used in Section 3.1 and 3.2 (The text of these two sections does not mention inversion. I assume that these are results from forward model simulations rather than inversion)? Or are they used for uncertainty estimation? If so, authors need to describe in the Method section how to compute the final uncertainty value from the ensemble inversions.

RESPONSE: We have changed "the posterior emissions were calculated for . . ." with "the posterior emissions are presented for . . .". We present the results of posterior BC emissions for the best representative species and the best prior dataset. We use all inversion BC emissions (using 12 BC species and 4 prior datasets) to calculate uncertainty, as stated in the next sentences clearly. Describing how the model ensemble was used is just 1-2 sentences and, given that it would stand as a separate section in the methodology, we have decided to simply mention it in the Abstract, in the last sentence of Introduction and describe it in section 3.3.

–The authors show that different wet removal parameters have little impact on simulated BC, which is interesting. But the authors can provide more information for readers to better evaluate this conclusion. (1) how BC aging and its hydrophilic/hydrophobic state is considered in the model? What is the uncertainty associated with this process? (2) what is the size of emitted BC, whether the size changes during transport, and is the simulation sensitive to the assumption of BC size? (3) are there evidence to support that the choices of parameter in Table 2 is adequate? (Grythe 2017 tested a larger ranges of parameters).

RESPONSE: (1) This has been already explained well in Grythe et al. paper referring to FLEXPART model. The level of detail of aerosol removal schemes in Lagrangian models is a limiting factor due to the Lagrangian model framework. A main consideration within this framework is that each transported computational particle is independent of others. Extensions of this concept to allow for non-linear chemistry exist or FLEXPART (Cassiani, 2013), but the reference version of FLEXPART is a purely linear transport model. Within such a linear model, it is impossible to include aerosol processes, which depend on the aerosol concentration (e.g., coagulation or non-linear chemical reactions). Furthermore, to facilitate consistency between forward and backward runs of FLEXPART, parameterizations that depend on the age of the aerosol (i.e. time after emission for primary aerosols) should be avoided as well. This limits the level of sophistication that can be incorporated into an aerosol removal scheme. Nevertheless, a realistic treatment of aerosols is possible even with these limitations.

In other words, the aerosol ageing processes are not readily included in FLEXPART and the constant removal parameters cannot account for this transformation. However, this may not cause significant uncertainties as there are several observations that prove that BC is transformed very quickly into particles with aged, hydrophylic characteristics (e.g. Wittbom et al., 2014).

(2) In section 2.2 we state that we have used a logarithmic size distribution with an aerodynamic mean diameter of 0.25 μm, a logarithmic standard deviation of 0.3 and a particle density of 1500 kg m$-$3. We also give a reference (Long et al., 2013). The reference explains very well the morphology and size of BC particles emitted in different steps (initial, after agglomeration, etc.). The change of the size during transport is accounted for using the logarithmic size distribution. Of course the simulation is sensitive to the size of particles, since changing the size distribution would also change the residence times of BC particles. However, we only examined the sensitivity to the new scavenging scheme that is used in FLEXPART and not to different particle sizes.

(3) The selection of parameters in Table 2 was done according to Grythe et al. (2017) paper. Like the reviewer says, Grythe et al. used a larger range of parameters. In our paper, when changing scavenging parameters of BC, except for conducting a sensitivity analysis and estimate uncertainties, our top priority was to select the best representa-

tive species for BC, hence scavenging coefficients should be realistic. Of course, if we change these parameters drastically (like in Grythe et al. paper), the variability of the posterior concentrations will increase. Though, this has been already done in Grythe et al. paper and, besides, we would not have realistic values anymore.

Cassiani, M.: The volumetric particle approach for concentration fluctuations and chemical reactions in Lagrangian particle and particle-grid models, Bound.-Lay. Meterol., 146, 207–233, 2013.

Wittbom, C., Eriksson, A. C., Rissler, J., Carlsson, J. E., Roldin, P., Nordin, E. Z., Nilsson, P. T., Swietlicki, E., Pagels, J. H., and Svenningsson, B.: Cloud droplet activity changes of soot aerosol upon smog chamber ageing, Atmos. Chem. Phys., 14, 9831–9854, doi:10.5194/acp-14-9831-2014, 2014.

Long, C. M., Nascarella, M. A. and Valberg, P. A.: Carbon black vs. black carbon and other airborne materials containing elemental carbon: Physical and chemical distinctions, Environ. Pollut., 181, 271–286, doi:10.1016/j.envpol.2013.06.009, 2013.

–The uncertainty estimates do not account for model transport errors and instrument errors. Section 2.2 and Figure 1 (g)-(i) show that the footprint varies greatly spatially. One may wonder how does this affect the inversion uncertainty? Can transport errors be more important than the choice of prior emission inventory and scavenging parameters in some regions distant from surface sites? Are observations informative for these regions? One of the major finding is gas flaring emissions from Russia but Figure 1 (g)-(i) show small footprint in these regions. Also, Figure 12 shows that locations of intensive gas flaring are shifted from prior inventory but Figure 1 (d)-(f) shows the inversion grid there is very coarse. It is worthwhile to check the posterior error covariance matrix to see how confident the inversion is about the conclusions?

RESPONSE: Model transport errors are not explicitly accounted for, but they are likely small compared to the error in the scavenging. We account for the scavenging error by running ensembles. And using e.g. inter-quartile range or SD over the ensemble to estimate your posterior uncertainty, then that already answers the question about how this affects the inversion uncertainty. As regards to the instrument errors, if the reviewer talks about measurements of BC, then these errors are included in the inversion. Each continuous measurement site gives concentrations plus measurement errors (see also previous comment).

Returning to scavenging errors, they are much more important that transport errors. This has been proved in Thompson et al. (2017) for the CH4 inversion in high northern latitudes. There, GFS versus ECMWF meteorology are compared as a proxy for transport error and is found transport error to be relatively small in most cases (note this does not calculate the true transport error but gives some indication). In the attached figure R2_F1, we show the error (in concentration units) calculated from all runs with different scavenging parameters for year 2013.

To answer the question about whether the observations are informative for remote sites, we can simply look at the footprints of Fig.1 in the manuscript for these observations. There, we can observe problems in 2014 in N. America due to the lack of measurements (already discussed in the paper) and over areas between Finland and Tiksi station (also discussed in the paper) for all years due to the same reason (lack of measurements).

Thompson, R. L., Sasakawa, M., Machida, T., Aalto, T., Worthy, D., Lavric, J. V., Lund Myhre, C., and Stohl, A.: Methane fluxes in the high northern latitudes for 2005–2013 estimated using a Bayesian atmospheric inversion, Atmos. Chem. Phys., 17, 3553-3572, https://doi.org/10.5194/acp-17-3553-2017, 2017

–Minor comments Abstract.

–The writing of the abstract can be improved. Line 17-19 and 23-25 seem to be out of place.

RESPONSE: Corrected. The first sentence (L17-19) was transferred to the next paragraph where the main results are summarized and the second one (L23-25), which is rather methodology-related, was transferred to the first paragraph (see manuscript with Track Changes).

–Line 37: main of atmospheric particular matter (?)

RESPONSE: Obviously a word is missing here, so we have corrected the sentence to: "Light absorbing species, such as black carbon (BC), are the main components of atmospheric particulate matter, affecting air quality, weather and climate." Please see manuscript with Track Changes.

–Line 39: gas flaring is open high- temperature combustion of natural gas in the oil/gas field. This source may be worth mentioning here because relevance to the results.

RESPONSE: We agree that it is a very good idea to also include this source due to its relevance with the results. Please see section 1. Introduction in the manuscript with Track Changes.

–Line 80: maybe worthwhile to compare the results with this emission dataset?

RESPONSE: The emissions dataset used in Popovicheva et al. paper is ECLIPSEv5 with GFED4, exactly the same with the ones used with the Bayesian inversion in the current paper.

–Line 152: the removal efficiency depends on BC aging (hydrophilic/hydrophobic) and size. How are they treated in the particle model? RESPONSE: See also previous comment on aging. Aerosol ageing processes are not readily included in FLEXPART and the constant removal parameters cannot account for this transformation. However, this may not cause significant uncertainties as there are several observations that prove that BC is transformed very quickly into particles with aged, hydrophylic characteristics (e.g. Wittbom et al., 2014).

Wittbom, C., Eriksson, A. C., Rissler, J., Carlsson, J. E., Roldin, P., Nordin, E. Z., Nilsson, P. T., Swietlicki, E., Pagels, J. H., and Svenningsson, B.: Cloud droplet activity changes of soot aerosol upon smog chamber ageing, Atmos. Chem. Phys., 14, 9831–9854, doi:10.5194/acp-14-9831-2014, 2014.

–Line 363-366: not clear how these uncertainties are individually computed and then combined. Equations are useful here. And it may also be a good idea to move the description in Method sections.

RESPONSE: We have clarified this now in section 3.3. Usually, one can combine two uncertainties from different processes by propagation and there are different methods to do this (see for example: http://lectureonline.cl.msu.edu/∼mmp/labs/error/e2.htm).

–Line 467-469: This type of uncertainty is not included in current uncertainty estimation but should be available by analyzing posterior error covariance matrix if B and R are properly assigned.

RESPONSE: Since we used an ensemble to calculate the posterior uncertainty, and take also into account the uncertainty due to scavenging, we do not have the error correlations. However, we can see the uncertainty for each grid cell in Whitehorse that is shown to be very high in 2013 and 2015, but not in 2014 (Fig.S5). If we also check the total footprint for all stations in 2014 (Fig. 1), we can easily understand that the coverage for this region is very poor. And this poor coverage is just due to the lack of measurements in this particular year, as we state in the manuscript.

–Section 4.4: The inversed emission seasonality is very interesting. I'd suggest summarizing the major finding about the seasonality in the abstract.

RESPONSE: Corrected! A sentence with the most important findings was introduced in Abstract (see manuscript with Track Changes).

–Line 553-555: what type of errors in the inversion?

RESPONSE: We agree with the reviewer. We do not really know what types of errors we have here. We have removed this comment. Please see last paragraph of section 4.4 in manuscript with Track Changes.

–Table 3: “four different optimized emission estimates are given”? Are they prior or posterior emissions? I am confused.

RESPONSE: We acknowledge the reviewer for pointing out this typo error. We have corrected the legend.

In this Table, we present annual posterior emissions of BC for N. America, N. Europe, N. Siberia and the 2 flaring regions in Russia. They are highlighted as “Posterior (ECLIPSEv5)”, because they were calculated using best representative species and best prior inventory. For comparison, we give the respective emissions for the same 5 regions in the four prior datasets highlighted as “ACCMIPv5 (prior)”, “EDGAR_HTAPv2.2 (prior)”, “MACCity (prior)” and “ECLIPSEv5 (prior)”.

–Figure 1: It’s impossible to read labels in panels (a)-(c). It may be a good idea to use the projection in Figure 6 for reader to compare different figures easily.

RESPONSE: The real goal of (a) – (c) panels there is to just show the density of the measurement stations that were used each year. All the characteristics, locations and instrument types can be seen in Table 1. Switching the projection to Mercator as suggested by the reviewer does not improve or solve the problem (see attached Figure R2_F2), so we would like to keep the current projection, which is consistent with the rest of the panels in Fig. 1

–Figure 2: Use a consistent projection for panels (a)-(g) and Figure 6 so readers can compare easily.

RESPONSE: We see the point of the reviewer, although that’s the main reason that we also present the difference between posterior and prior emissions in Figure 6; so the readers can compare easily. We have corrected Figure 2 as suggested, although we doubt that one will be able to distinguish different source locations on such a complex Figure.

–Figure 3 and 4: Two figures provide very similar information. Consider removing one.

RESPONSE: These 2 different pictures are used for different reasons in the text. They also use 2 different statistics and having 2 statistics is always better that having only one. Since there are no space limitations in ACP, we would prefer to keep both of them as an additional evidence of our findings within the text.

–Figure 7 - 9: These evaluations are quite interesting. But since the inversion is done with ECLIPSEv5, it is a fair comparison between posterior emissions with averaged RMSE from four emission inventories. RMSEpri from ECLIPSEv5 should be listed as well.

RESPONSE: Corrected!! We have introduced RMSE values for ECLIPSE as well, as suggested by the reviewer.

–Figure 9: The difference between posterior and ECLIPSEv5 is very small, which may be indicative that the observations are not informative to emissions in this region. See major comments for uncertainty estimations.

RESPONSE: We agree with the reviewer that lack of observations there constrains sources poorly in areas between Finland up to Tiksi station pushing the inversion towards prior emissions. However, there is no solution for this.

–Figure 11-12: "Black rectangles show vegetation fires adopted from Hao et al." Choose another color. Black color is already used for map borders. Also, they are better referred to as dots rather than rectangles.

RESPONSE: Corrected! We have introduced dark green points to represent fire spots. See Track Changes manuscript.

–Figure 13: The prior emissions plotted in panel d (N.Siberia) of Figure 13 seems to be wrong as they are much lower than the annual total emissions reported in Table 3.

RESPONSE: Again, thanks a lot for pointing out this mistake. We had accidentally plotted another area, where N. Siberia should be. We have now corrected it! Please see Track Changes.

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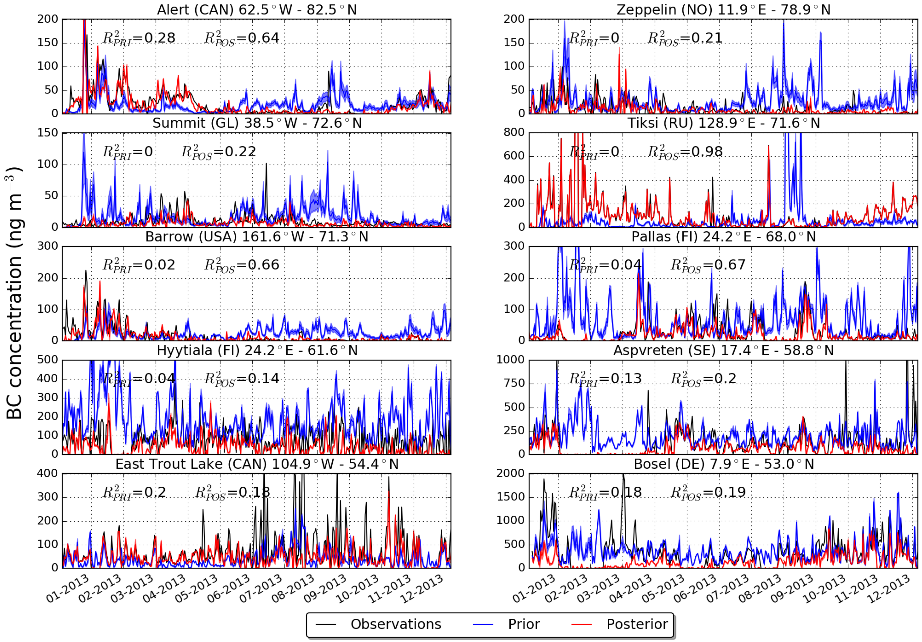

Fig. 1. Timeseries of surface concentrations and associated uncertainties in 2013

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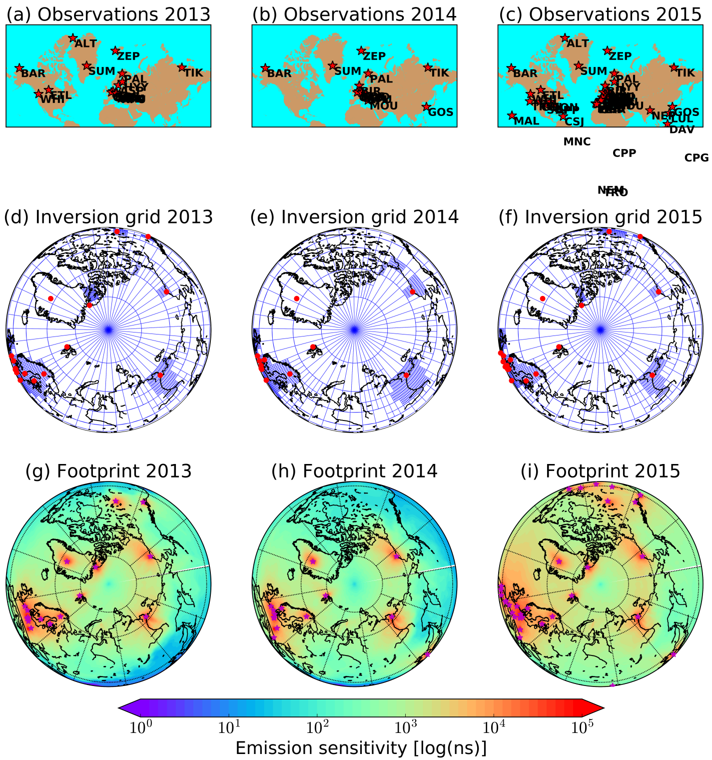

Fig. 2. Manuscript Figure 1 in different projection

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