Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2017-357-AC2, 2017

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Do contemporary (1980–2015) emissions determine the elemental carbon deposition trend at Holtedahlfonna glacier, Svalbard?" *by* Meri M. Ruppel et al.

Meri M. Ruppel et al.

meri.ruppel@helsinki.fi

Received and published: 1 September 2017

Dear Anonymous Referee #2,

We are grateful for your efforts and overall positive evaluation of our manuscript, and the constructive comments that have helped us to further improve the paper. We find your comments well-justified and have revised the manuscript accordingly. Below we give our detailed responses to your comments and describe the revisions prepared for the manuscript. The Referee comments are cited with REFEREE 2 and our responses in regular type while revisions prepared to the manuscript are marked with quotation marks.

REFEREE 2: Applying the EUSAAR_2 protocol to EC measurements in liquid phase sample is not straightforward, as eg the sample needs to be filtered, and that the efficiency of filters to capture EC can be limited (Eg Torres et al., 2014; Lim et al., 2014). Lim et al. (AMT, 2014) have reported that the filtration efficiency (ie the amount of EC retained on a filter) can be as low as 20% for small EC particle diameters (eg 100 nm MED). I am wondering if such artifact might partially explain the fact that modeled BC deposition are higher than observed EC deposition. Overall, any information about the size distributions of BC in snow/ice would be interesting, as larger particles could drive larger observed EC concentrations (and seasonal melt at the surface of the glacier can promote larger BC particles by aggregation). Considering the challenge of measuring BC in snow, combining results from different analytical methods would be more solid. If additional measurements are not possible (eg involving an SP2 analyzer) for this study, the manuscript should at least include more discussion about potential uncertainties related to the analytical method. I understand that discussion on that topic is included in the 2014 Ruppel paper, but I would recommend at least to refer more clearly to it.

The reviewer highlights important points and we agree with these uncertainties in the filter based EC measurements. Undercatchment of small EC particles is a known error source in these filter based EC measurements, causing a ca. 22 % underestimation of EC concentrations in the used set-up (Forsström et al., 2013). We agree that we should discuss this issue more, and have subsequently added this information to Section 2.2. and made clearer references to the Ruppel et al. (2014) paper where this issue has been discussed in more detail.

P4, L32-39: "The used methodology includes uncertainties that are described in more detail in Ruppel et al. (2014). In short, in liquid samples smallest EC particles may go through the filter leading to a quantified undercatchment of ca. 22 % for the used filtering set-up (Forsström et al., 2013). In addition, from each filter sample (11.34 cm-2) only a smaller punch (1.5 cm-2) is analysed for EC. To evaluate the uncertainties

caused by this subsampling, triplicate analyses were prepared for five ice core samples. These measurements (Fir. 4 c) showed an average relative standard deviation of 8.5 % (range of relative standard deviation = 5.3–13.7 %) that is smaller than reported e.g. in Ruppel et al. (2014). Combined (added together in quadrature) these error sources cause a ca. 23.6 % uncertainty in our current EC measurements."

However, the reviewer mentions by mistake that the filtrations inefficiency may potentially cause the observed EC deposition to be lower than the modelled BC deposition, while in fact it is the other way around: the observed EC deposition is significantly higher than the modelled BC deposition. The fact that observed and modelled BC concentrations and deposition very often deviate significantly from one another (e.g. Koch et al., 2009, 2011) is mostly not related to uncertainties in the observational measurements but the model parameterization (e.g. sizes of BC particles and their definition potentially deviating from what has been measured, emissions, transportation and ageing of BC particles etc.). We have added discussed model limitations and why these will affect the modelled BC values on P 12, L 16-31, also according to the suggestions of Referee #1.

We agree with the reviewer that considering the challenges in BC measurements from snow it would be most solid to combine results from different analytical methods. Unfortunately, the SP2 methodology was not available for this study but efforts are taken to make such measurements in future studies.

REFEREE 2: The EC data should include quantified uncertainties.

Yes, we agree. Quantified uncertainties have been added in Figure 4c for the ice core measurements. In addition, we have included some new text on the issue on P 4, L 32-39, as cited above.

REFEREE 2: The paper misses a direct comparison between model atmospheric BC and direct atmospheric observations, eg from Ny Alesund Station. This would support more clearly the model outputs (as only suggested in the manuscript).

Thank you, this is a justified concern that was also raised by the other referee. Accordingly, a new Figure (5) comparing observed and modelled annual average and monthly atmospheric BC concentrations at Ny-Ålesund has been added, together with two paragraphs in Section 3.3. for model validation (P7, L 4-19). For more details see responses to Referee #1.

REFEREE 2: I understand that observed EC deposition does not corroborate directly with globalscale emission patterns. Can we learn more by considering regional emissions patterns? The emissions description seems to miss details about such regional patterns, and their relative impacts at the study site.

This is a justified comment by the reviewer. We have included the emission trends of 40° N in the manuscript because this region is considered a significant source region for BC deposited in the Arctic (e.g. AMAP 2011). It is also shown in several back-trajectory modeling studies that northern Russia is a strong (or even dominant) source region for BC arriving in Svalbard (e.g. Hirdman et al., 2010 and references therein; Stohl et al., 2013; Winiger et al., 2015). Therefore, it could be meaningful to compare the BC concentration and deposition trends at Holtedahlfonna with Russian BC emissions. However, several studies have implicated that the BC emission inventories from Russia have not been reliable or up to date (e.g. Stohl et al., 2013; Huang et al., 2015). Huang et al. (2015) published the first regional scale emission inventory for Russia showing severe underestimation in total BC emissions, particularly from flaring, and also miss-allocations of emission sources. Furthermore, the up-dated emission inventory was published only for 2010. Consequently, it would not be meaningful to attempt any deeper comparison of regional emission patterns with our current data, particularly as the temporal trend of up-dated emission inventories would not be possible.

One of the objectives of this manuscript was to assess whether flaring or any other individual emission source could have been responsible for the EC deposition increase observed at Holtedahlfonna from 1970 to 2004. For this, it was important to use consistent emission data available for the whole study period, although it may not have

been the most up-to-date or accurate. In future, it would surely be fruitful to assess the emissions on a more regional scale but this was not in the scope of the current paper. Furthermore, our own study shows the limitations of chemical transport models to capture regional details such as the seasonal variation in BC at a remote location. This is why our study focuses on trends over several decades. Other models, such as the trajectory models (e.g. Grythe et al., 2017), would probably be able to better tackle this particular research question of how regional emission patterns affect our Svalbard study site. For clarity, we have added information on the source areas relevant for the study site in the revised manuscript, while we have not included new discussion on regional emission patterns affecting the study site due to reasons listed above.

P5, L26-27: "Generally, BC emissions north of 40° N are considered significant for the Arctic (AMAP, 2011)."

P5, L34-38: "The total global BC emissions have increased in the study period while north of 40° N they have decreased (Fig. 3). Svalbard receives atmospheric transportation dominantly from Eurasia (e.g. AMAP, 2011), and anthropogenic BC emissions from this region have decreased in the study period while natural fire emissions have increased (e.g. Bond et al., 2007; Lamarque et al., 2010)."

Clarification: P11, L8-9: "Notably, however, the modelled annual BC deposition does not clearly follow (or correlate to) the declining north of 40° N BC emissions (Fig. 3b) or modelled and measured atmospheric BC concentration trends (Fig. 6)."

REFEREE 2: P5-L35 : typo "llike"

Thanks, corrected.

REFEREE 2: P11-l7 typo "second may"

Thanks, corrected.

References:

AMAP: The Impact of Black Carbon on Arctic Climate (2011). Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway, 72 pp, 2011.

Forsström, S., Isaksson, E., Skeie, R. B., Ström, J., Pedersen, C. A., Hudson, S. R., Berntsen, T. K., Lihavainen, H., Godtliebsen, F., and Gerland, S.: Elemental carbon measurements in European Arctic snow packs, J. Geophys. Res. -Atmos., 118, 13614–13627, doi:10.1002/2013JD019886, 2013.

Grythe, H., Kristiansen, N. I., Zwaaftink, C. D. G., Eckhardt, S., Strom, J., Tunved, P., Krejci, R., and Stohl, A.: A new aerosol wet removal scheme for the Lagrangian particle model FLEXPART v10, Geosci. Model Dev., 10, 1447-1466, DOI: 10.5194/gmd-10-1447-2017, 2017.

Koch, D., Schulz, M., Kinne, S., McNaughton, C., Spackman, J. R., Balkanski, Y., Bauer, S., Berntsen, T., Bond, T. C., Boucher, O., Chin, M., Clarke, A., De Luca, N., Dentener, F., Diehl, T., Dubovik, O., Easter, R., Fahey, D. W., Feichter, J., Fillmore, D., Freitag, S., Ghan, S., Ginoux, P., Gong, S., Horowitz, L., Iversen, T., Kirkevåg, A., Klimont, Z., Kondo, Y., Krol, M., Liu, X., Miller, R., Montanaro, V., Moteki, N., Myhre, G., Penner, J. E., Perlwitz, J., Pitari, G., Reddy, S., Sahu, L., Sakamoto, H., Schuster, G., Schwarz, J. P., Seland, Ø., Stier, P., Takegawa, N., Takemura, T., Textor, C., van Aardenne, J. A., and Zhao, Y.: Evaluation of black carbon estimations in global aerosol models, Atmos. Chem. Phys., 9, 9001–9026, https://doi.org/10.5194/acp-9-9001-2009, 2009.

Hirdman, D., Sodemann, H., Eckhardt, S., Burkhart, J. F., Jefferson, A., Mefford, T., Quinn, P. K., Sharma, S., Ström, J., and Stohl, A.: Source identification of short-lived air pollutants in the Arctic using statistical analysis of measurement data and particle dispersion model output, Atmos. Chem. Phys., 10, 669–693, doi:10.5194/acp-10-669-2010, 2010.

Huang, K., Fu, J. S., Prikhodko, V. Y., Storey, J. M., Romanov, A., Hodson, E. L., Cresko, J., Morozova, I., Ignatieva, Y., and Cabaniss, J.: Russian anthropogenic black

carbon: Emission reconstruction and Arctic black carbon simulation, J. Geophys. Res. Atmos., 120, doi:10.1002/2015JD023358, 2015.

Koch, D., Bauer, S., Del Genio, A., Faluvegi, G., McConnell, J. R., Menon, S., Miller, R. L., Rind, D., Ruedy, R., Schmidt, G. A., Shindel, D.: Coupled aerosol-chemistry-climate twentieth century transient model investigation: Trends in short-lived species and climate responses, J. Climate, 24, 2693–2714, doi:10.1175/2011JCLI3582.1, 2011.

Ruppel, M. M., Isaksson, I., Ström, J., Beaudon, E., Svensson, J., Pedersen, C. A., and Korhola, A.: Increase in elemental carbon values between 1970 and 2004 observed in a 300-year ice core from Holtedahlfonna (Svalbard), Atmos. Chem. Phys., 14, 11447–11460, doi:10.5194/acp-14-11447-2014, 2014.

Stohl, A., Klimont, Z., Eckhardt, S., Kupiainen, K., Shevchenko, V. P., Kopeikin, V. M., and Novigatsky, A. N.: Black carbon in the Arctic: the underestimated role of gas flaring and residential combustion emissions, Atmos. Chem. Phys., 13, 8833–8855, doi:10.5194/acp-13-8833-2013, 2013.

Winiger, P, Andersson, A., Eckhardt, S., Stohl, A., Semiletov, I. P., Dudarev, O. V., Charkin, A, Shakhova, N., Klimont, Z., Heyes, C., and Gustafsson, Ö.: Siberian Arctic black carbon sources constrained by model and observation, Proc. Nat. Sci. Acad. USA, 114, E1054–E1061, doi: 10.1073/pnas.1613401114, 2017.

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-357, 2017.