

Here we reply to each comment given by Anonymous Referee #2. The comments of the referee are given in italics and our replies in regular type.

## **Anonymous Referee #2**

*Ruppel et al analyzed a Svalbard ice for elemental carbon using a thermal optical method. Overall this paper contributes to our understanding of temporal variations in BC deposition in the Arctic, and provides valuable discussion of the factors controlling BC deposition at Svalbard. Detailed comments are provided below, including suggestions for revisions to the manuscript to clarify the interpretation of the record and more fully acknowledge uncertainties in the analytical technique.*

*Title should omit 'Unexpected' and preferably refer to the year that the increase started rather than 'last 30 years.' Since the core was drilled in 2005, the last 30 years can be misinterpreted. As discussed below, it is misleading to state that there has been an increase since 1970.*

We mostly agree with the reviewer. The suggested title was somewhat vague, and we will omit the 'Unexpected'. However, we partly disagree with the reviewer that the increase in EC values would not have started in 1970 (discussed more thoroughly below). Now we suggest the title:

"Increase in elemental carbon values between 1970 and 2004 observed in a 300-year ice core from Holtedahlfonna (Svalbard)"

### *Abstract:*

*See comments below claiming increase since 1970, and fix accordingly.*

The reviewer makes a fair point that our statement about increased EC values since 1970 could be misleading in the context of the trend in the whole 300-year ice core record. We will discuss the issue more thoroughly below. However, we will clarify our statement also in the abstract.

*l. 15 Sentence starting 'Several hypotheses' is difficult to follow and needs revision.*

We will clarify this statement in the revised version of the manuscript.

*l. 22 omit the in 'in the recent decades'. This should be changed however as referred to in the title comment.*

We will clarify this statement in the revised version of the manuscript.

### *Introduction:*

*Writing in opening sentence can be improved (fix 'which has been suggested to be explained by changes: : :')*

Will be done.

*p. 13200 l27 expand on statement that Svalbard glaciers are expected to have a different source attribution than Greenland.*

We agree that this may have been an unclear statement. It is stated several times in later parts of the manuscript that based on back-trajectory modelling Greenland receives most of its airmasses and pollution deposition from North America whereas Svalbard is more susceptible for emissions in (northern) Europe and Russia (e.g. p. 13210, l. 7-16). For clarity, we will rephrase the statement in question in the revised version of the manuscript.

### 2.3 Uncertainties

*p13204 l6 The discussion on the filter efficiency of the filters is misleading in that it leads the reader to think that there was no filter efficiency issue. Torres 2014 showed that there was undercatch using quartz fiber filters, but that filtration could be greatly improved with the addition of salts. I've tested this in my own laboratory and have found that filtration of snow and ice samples using quartz fiber filters is improved considerably when using salts. Additionally when using thermal optical analyses we use three filters in line (separated, not stacked on top of one another) following the approach of Odelle Hadley that also improves filter efficiency. While Schwarz et al 2013 shows that BC particles in snow can shift to larger size particles, considerable mass of BC in snow is below .5  $\mu\text{m}$ . Schwarz also demonstrates that the mass absorption cross section of smaller BC particles is much greater than for large particles (see Fig 1 of Schwarz 2013), so if albedo is of interest (which it is for this study), the smaller particles, which aren't being filtered efficiently, are certainly of interest. Ideally the authors would quantify the EC not captured on the filters. At a minimum this section needs revising to fully disclose these issues.*

The reviewer makes several fair points. We will revise the paragraph in question according to the comments.

First, we will clarify that despite the presence of bigger BC particles in snow and ice than in the atmosphere, the used method and filters undercatch some of the smallest BC particles. This will result in the presented EC concentrations likely being an underestimate of real EC concentrations. Forsström et al. (2013) reported this bias to be 22% on average by using the same filters as in this study and by analysing a second filter (Nuclepore 0.4  $\mu\text{m}$ ) in line with the ISSW (Integrating Sphere/Integrating Sandwich method) method used by Doherty et al. (2010; 2013) at the University of Washington. However, the amount of undercatch samples in Forsström et al. (2013) was not enough to confidently develop a quantitative correction of undercatch with the used method. But as the undercatch values reported in Forsström et al. (2013) are from snow samples partly collected from Svalbard, we feel that they could provide an estimate for the related error in our data.

Second, by the time the filtering of the ice core was done for this study (February 2013), we were unaware that the filtering efficiency could be increased with addition of salts as the study in question (Torres et al., 2014) had not yet been published. During our own testing, we have not noticed that filtering the samples in line, as suggested by the reviewer, would increase the filtering efficiency as no EC has been detected on the second filter in line with the thermal-optical analysis. This may also be a consequence of different types of quartz fibre filters used.

Third, we will include percentage values for the different possible biases present in our study. As mentioned in the earlier paragraphs of the uncertainty section of the manuscript, there is a general standard deviation in the EC results due to heterogeneous loading of EC on the filters. This bias is at most ca. 20% (although lower for most of the filters) as measured in the present and Svensson et al. (2013) studies. In addition, Forsström et al. (2013) reported the undercatch of the used filters to be ca. 22%. If these separate error sources are added together in quadrature, as in Schwarz et al. (2012), we end up with an estimate of maximum 35-40% total uncertainty related to our measurements.

Finally, we agree with the reviewer that the ultimate motivation for this work is the radiative impact of BC and that small particles are certainly of interest because they may have a greater mass absorption cross section than large particles in snow. However, the main message of our results is that there has been an increase of EC concentrations in snow at least at the ice coring site during 1970-2004 which is contrary to the general consensus of declining atmospheric Arctic BC concentrations since 1989. Even if this was only caused by an increase of larger BC particles with lower radiative impact than smaller BC particles, the increase is so significant that it must have had an impact on the radiative transfer at the ice coring site which is unexpected based on atmospheric measurements.

*Results and Discussion*

*p. 13205 I1 'are show clearly' fix writing.*

Will be done.

*p. 13206 I3 While from 1970 to the top of the record EC does increase, EC in the 1970s and 1980s is still within the range of concentrations earlier in the 20th century. It is only the very top of the record (1990s-2004) that EC exceeds concentrations earlier in the record. The discussion should be revised to not suggest that the 1970-1980s were anomalously high for the record. The same applies later in the record when EC deposition trends are discussed.*

This is a useful comment as it shows that we need to clarify some statements. At no point did we mean to suggest that the EC values of 1970s and 80s were anomalous for our record. However, based on available information from atmospheric measurements and the Greenland ice cores, our trend of increasing EC values from 1970 onwards is unexpected. Based on the available information we could have expected the EC values to stay stable or even decrease during the last decades of the record. While the concentrations at around 1910 and 1950 presented local maxima, the concentration since 1970 has increased more or less steady on a decadal perspective. Thereby we will clarify our statements in the revised version of the manuscript but we will keep our inference that EC values have increased since 1970, as this is the turning point of the record where an increase starts instead of expected stable or decreasing values.

*I. 5 It would be useful to plot the atmospheric BC records from 1989 along with the ice core record. It appears the ice core EC peaks around 2000, but is lower since then. Is the timing consistent between the records?*

We agree with the reviewer that the ice core record seems to peak around 2000. However, we do not think that the last three measurement points in the ice core should be seen as a trend to decreasing EC values since 2000 as these are statistically too few points to depict a trend. As mentioned in the text (p. 13212, I. 2-11) atmospheric measurements of BC concentrations are available from Ny Ålesund only since 1998. Consequently, the ice core record and atmospheric measurements could be compared to each other only for the last three ice-core samples. We do not think that such a comparison would be meaningful. Atmospheric measurements from Barrow and Alert are available from 1989 onwards but comparison to these records does not give much further information either. All atmospheric measurements in the Arctic have measured decreasing concentrations since the beginning of the measurements and show no peak around 2000. We will clarify this in the revised version of the manuscript, e.g. by citing a study by Sharma et al. (2013) showing an overall decline of 40% in atmospheric BC measurements in the Arctic from 1990 to 2009.

Secondly, as we discuss in the scavenging section (3.4) of the manuscript, the atmospheric measurements should not and cannot be directly compared to ice-core concentrations or deposition since ice-core values are influenced by several other atmospheric and post-depositional factors and not just atmospheric concentrations.

*I. 24 the discussion on trends from the European Alps would be easier to interpret if the studies were referred to by their locations rather than just authors.*

We will do this in the revised version of the manuscript.

*p. 13206 For dusty sites like the Himalayas the dust causes interference with the thermal optical method (e.g., see work by Wang Mo), causing additional problems comparing records.*

We will include this comment in the revised version of the manuscript.

*p. 13207 I.22 Xu 2012 showed that under conditions of strong melt that BC is enriched even more so over the superimposed ice layer than at the surface.*

We will include this comment in the revised version of the manuscript. Due to the annual to biannual resolution of our record during the 20<sup>th</sup> century, this fact does not affect our results.

*P13208 I. 10 I'm not following the logic of the interpretation here. Snow accumulation is stated to be lower during 1930-1960, but that isn't what the data in Fig 3c shows. If accumulation is higher during that period and EC was constant in the atmosphere, lower EC concentrations would be expected, but this isn't what the data shows. That EC is higher (Fig 3b) during the 1920-1970 period, which corresponds to a period of relatively higher snow accumulation would suggest that EC in the atmosphere was higher during this time.*

The reviewer fairly points to possibly unclear statements in the text. In this part of the manuscript we are specifically comparing EC concentration and deposition values. Generally, if atmospheric and post-depositional processes stay constant, concentrations and deposition present similar trends in ice cores. If concentration and deposition trends differ from each other, again generally, the most likely explanation is a change in snow accumulation. This change in snow accumulation may, under stable atmospheric processes, dilute or concentrate EC amounts in snow/ice whereas the deposition is not affected. The key point in this discussion is the comparison of the snow accumulation rate before and after the discrepancies between the concentration and deposition trend, not necessary the relative snow accumulation rate of the whole record. In our case, the snow accumulation rate during 1930-1960 was clearly lower than during 1910-1930 and 1960-2000. This concentrated the EC in the ice core, causing the concentrations to be higher around 1920-1970 than during adjacent time periods despite the deposition not increasing. It is the simplest explanation for the observed differences between the concentration and deposition records. We will clarify our reasoning in the revised version of the manuscript.

*I. 14 include plots of the Greenland ice core BC data in Fig 3. to compare timing between records.*

We will include the Greenland ice core BC data in the revised manuscript.

*13209 I. 8 why specifying N. America? If the majority of EC is thought to originate from N. America this would make sense, but this isn't stated. For this section it would be useful to plot the regional emission inventory data from Novakov and/or Bond. These are based on fossil fuel inventories, so some attention should also be given to the idea that the emission inventory estimated don't capture all sources.*

This is a fair point, and we will clarify the statements in the revised version of the manuscript. We mention North America since together with Europe and Russia it is believed to be the most important emission source for BC in the Arctic (as mentioned in other parts of the text). We will clarify that based on the emission inventories, at the same time with decreasing BC emissions from North America, European emissions stayed quite constant in the latter half of the 20<sup>th</sup> century. To visualize this we will include, in addition to the historical global BC emission data, the regional emission inventory data from North America, Europe and the former USSR based on Bond et al. (2007). We will also clarify that, as the reviewer points out, these emission inventories represent only emissions from fossil fuel sources and may also be limited even in their case, as, for instance, flaring has not necessarily sufficiently been taken into account in them (Stohl et al., 2013). Also biomass burning, such as forest fires in Russia and Canada, is expected to be significant source of BC in the Arctic which we should mention.

*p 13212 I15-21 The discussion on scavenging efficiency and its link to temp and precipitation needs to be referenced to the source of this information.*

The reference for this discussion is provided in the next sentence of the paragraph (Cozic et al., 2007).

*p. 13214 I17 same as discussion on increase in EC from 1970s. Stating that temp increased from 1960 is misleading since the temp record actually shows that temp in 1960 was cooler. Revise wording.*

We will revise the wording to clarify our statement. The summer temperatures started to increase in the 1960s after an initial cool phase.

*I. 22 awkward start to sentence. Omit 'Though.' The intensifying summer melt leads the EC concentration by numerous years, which should be made clear when discussing that the two records correspond. If the melt index isn't reliable as a melt index post 1990, the data shouldn't be presented for this section of the record. What did the visual record of the ice core show re: melt layers? Is there a record of melt layers from the core that can be included here?*

We will rephrase the sentence. We will also add the comment that the intensifying summer melt leads the EC concentration record by several years. However, we do not think that it is necessary to cut the melt index data so that we would not present the data post 1990. In the text we explain carefully why the melt index data fails after 1990 and omitting the data would be suspicious. We consider our explanation for the turn in the melt index to be valid.

The best information that we have on the melt layers in the ice core is presented in Beaudon et al. (2013) as the density data of the ice core (Beaudon et al., 2013, and Fig. 4 therein). It shows that the density of the ice core generally decreases since 1970 towards the core top. The lower density at the top is a common feature of ice cores and is explained by the top of the core being firn not yet compacted to ice by the weight of the overlying snow and firn. However, dense melt layers are visible also in the firn section of the ice core between 1970 and 2004 (see density data in Beaudon et al., 2013). We will make sure that we clearly discuss the presence of melt layers at the top of the ice core and refer to the

data available in Beaudon et al. (2013) in the revised version of the manuscript. We do not believe that the addition of the density data is necessary for this manuscript.

*p. 13215 l. 15 Xu 2012 showed that with high amounts of melt the BC can move through the snowpack.*

We can note in the revised version of the manuscript that BC may percolate to some extent in the annual snow pack, as also shown by Doherty et al. (2013). However, BC does not percolate to deeper layers through the annual melt layer. Therefore, this percolation does not affect our results which are given in annual to biannual resolution for the 20<sup>th</sup> century.

*l. 28. Also importantly during the summer greater dry deposition would occur which would also lead to higher concentrations.*

A fair comment – we can include this comment in the revised manuscript.

*p. 13216 l. 7 Recommend saying it differs from rather than contradicts. As stated above, more attention should be given to the regional changes in emissions for comparison with the ice core record.*

The recommendation for wording is good. We will discuss more thoroughly the regional (northern hemisphere, mid to high latitude) changes in BC emissions for comparison with the ice core record, by including some regional plots (emissions from Europe, North America and Former USSR). At the same time we want to stress that, in addition of not presenting biomass sources for BC, these emission inventories (e.g. Bond et al., 2007) may insufficiently portray all fossil fuel sources. For instance, Russian flaring has not been sufficiently accounted for (or is neglected) in the inventories, and as a high-latitude emission source it may have a key influence on Arctic BC concentrations and deposition (Stohl et al., 2013).

*p. 13216 I don't expect that the difference in the trends between Greenland and Svalbard are related to the analytical methods. Different trends were observed at cores near each other in the Himalayas likely due to differences in analytical techniques, but this is likely because of the high dust concentrations in snow and ice in this regions that cause interference with the thermal-optical method. For relatively clean Arctic cores I don't think the trends would differ much due to analytical differences (however the measured concentrations will differ).*

Again, the reviewer makes a fair point. It is true that the differing results from the Himalayas between methods may partly be caused by dust interfering with the thermal-optical method, while dust should not be a problem with Arctic ice cores. However, as we discuss, the different results acquired by the SP2 and thermal-optical methods from Arctic ice cores is probably partly caused by neither of these methods being able to detect the whole spectrum of different-sized BC particles present in the snow and ice. The SP2 method will not measure larger BC particles present in snow and especially ice while the thermal-optical method will likely undercatch smallest BC particles. This will inevitably result in the methods measuring different BC concentrations and possibly even magnitudes of concentrations. If the proportion of smaller to bigger BC particles in the ice cores stays constant throughout the whole records (temporally), then the results given by the different methods will only differ in BC concentration. However, if the size distribution of BC present in Arctic ice cores has varied through time, the temporal trends of BC concentrations in the Arctic ice cores measured by the different methods can vary. For instance, BC particles deposited since ca.

1970 might have increased in size, e.g. as a consequence of new BC sources or possibly due to atmospheric and/or post-depositional processes increasing the size distribution of BC particles found in snow. Such processes would have resulted in the bigger particles being captured on Svalbard with the thermal-optical method while the SP2 method used on the Greenland ice cores would not have been able to measure these. Alternatively, atmospheric and post-depositional processes increasing the size of BC particles deposited in the ice cores may have been present in Svalbard but not Greenland (e.g. more liquid water clouds causing increased scavenging efficiency and more summer melt on Svalbard in comparison to Greenland). We believe that these issues may partly be responsible for the different trends observed in the Greenland and Svalbard ice cores analysed with different methods. We will clarify this in the revised version of the manuscript. However, this has not yet been tested in detail (but will be in future work by the authors) and thus for the time being remains one hypothesis among the others.