

Reply to referee comments given on Svensson et al., The Cryosphere Discussions, 2015

Here we present point-by-point responses to the referee comments given on the manuscript “Soot on snow experiments: light-absorbing impurities effect on the natural snowpack.” Comments from the referees are given in black plain text in this document and our response is indicated in blue color.

The consequent changes made according to the referee comments have greatly improved the revised manuscript. Deletions in manuscript have been marked by ~~red-strikethrough~~, whereas additions to the paper are indicated by blue color.

Sincerely, on behalf of my coauthors,

Jonas Svensson

Anonymous Referee #1

Svensson et al. report on the impact of black carbon (BC) in snow on the albedo and the melting of the snowpack. They performed experiments in the field, during which they artificially caused the deposition of additional absorbing impurities (including an overwhelming fraction of BC) on natural snowpack. The experiments were performed with different set-ups during three winters (2011, 2012, 2013) at three different sites in Finland. The presented results concern measurements of BC concentrations in the snow and how they evolved after the BC deposition until the melting of the snow, albedo measurements at undisturbed and affected sites compared to simulations with the SNICAR model, and observations of physical snowpack properties related to the experiments. The authors claim that in general albedo measurements and simulations agree. Further conclusions consist of a set of recommendations for further field experiments concerning the impact of BC and other absorbers on snow albedo and further processes. BC in snow has recently attracted strong scientific interest due to its potential to lower the snow albedo impacting metamorphism, snow melting, and radiative forcing. Global, regional, and local model studies based on radiative transfer theories applied to snow demonstrated the potential impact of BC in snow in regions like the Arctic or the Himalayas even with the relatively low BC in snow concentrations encountered in these regions. However, dedicated field experiments based on manipulating BC concentrations in real snow to quantify the link between BC and albedo and snow melting are very limited. Therefore, any new measurements to study these links are highly welcome especially during the melting period of the snowpack. Unfortunately, the presented experiments and the analysis seem to be seriously flawed so that sound conclusions are either limited or even impossible. As a result, the novelty of the presented results is rather limited. In the comments below, I describe a number of my concerns regarding the manuscript. I can only recommend refusing the publication of the manuscript in the Cryosphere.

Comments

Chapter 3.1: The authors discuss in this chapter the evolution of BC in the snow during the SoS2013 experiments. For example, because the concentrations decreased from 1465 to 529 ppb between 8 and 17 April they state that only 36 % of the initial soot particles were observed at the surface after 9 days. Such a reasoning based on observed concentrations is wrong. The calculation can only be based on budgets of BC in the snow, not on concentrations. The authors go even further to calculate a sum of observed concentrations (see Table 2, for example 746 as a sum of the concentrations observed in five snow layers at spot 7 on 17 April). Such a sum is useless because it depends on the number of samples. For instance, it would be a factor of 2 higher if the authors had sampled 10 layers. The authors continue to calculate the fraction in the different layers based on the concentrations and the sum of the concentrations. This gives more or less correct fractions only if the entire snowpack is represented by exactly the five sampled layers. It must also be assumed that the density variation inside the snowpack is negligible, but it remains unclear if that was the case in all sampled snow pits. Finally, a direct comparison of the concentration in a top layer, which is in one case 7 cm thick (8 April), with the concentration in the top layer of the subsequent snow pits, which is 5 cm thick (17 and 24 April), is misleading if the different thicknesses are not considered. This is even more important in these experiments, where a strong BC gradient between the surface and the underlying layers can be expected due to the design of the BC deposition experiments.

Chapter 3.1: To derive reliable data on the BC in snow trends several steps are needed. 1. The background BC in snow concentrations for the entire snow columns need to be measured. The authors give an average BC in snow concentrations for SoS2011. Is this only surface snow or the entire column? But no background data for SoS2013 are given except maybe the entry "9B (reference)" without any further description in the text. 2. BC budgets for all sampled layers need to be calculated. This requires BC concentration, BC background concentrations, and snow density measurements. However, it appears that at the start of the SoS2013 experiments only the surface snow layer was sampled. 3. Total BC in snow columns can be calculated by summing up the budgets of the individual layer and taking into account the full snow column. According to the

manuscript it seems as if the authors do not have all necessary data to perform such calculations. If that is the case, their results remain qualitative and appears impossible to derive scientifically sound numbers.

This issue has been correctly pointed out by the referee and the EC concentrations were incorrectly used in the manuscript. In order to perform the suggested calculations some data is lacking. Therefore, we have decided to change our focus and discussion on the BC in snow for these experiments. We will now shift away from the vertical movement of the BC particles in the snowpack (and that discussion). Instead the emphasis will be on the surface layers where the data is sufficient and when necessary only give a qualitative approach to the results and discussion. This fits into the additional changes that are done to the revised manuscript, where we are changing the focus to the albedo (broadband) and the effects on it caused by the soot deposited on the snow surface (i.e. less focus on the effects from the soot on the snow properties).

Change in the manuscript:

Table 2 has been removed from the older version of the manuscript. Section 3.1 has been rewritten with an emphasis on the surface layer (applicable to the EC concentrations in the snow; other sections connected to the focus shift will be highlighted elsewhere in this document).

Chapter 3.4: The authors perform a linear regression using their observations as shown in Fig. 4. How is this justified if the widely accepted radiative transfer theory predicts a non-linear behavior between BC concentrations and albedo?

The regression used in Fig. 4 was not linear, it was a logarithm of the EC concentration. Using this approach it ends up being linear, however. In the revised manuscript we redid a fitting to our experimental data. We also added another fitting to the data, which is based on the albedo over the first few days compared to only the first day in the manuscript. This is to present alternative numbers and to show that the fitting is more robust over this period of time.

Change in the manuscript:

A new non-linear fit is show in Fig. 7, and further discussed in section 3.4.

Chapter 4: The authors list seven recommendations for future studies regarding BC in snow experiments. In my opinion, at least five of these seven recommendations (1. experiments with low BC in snow concentrations are needed; 4. experiments over longer periods needed; 5. more detailed measurements are needed to follow changes in the snowpack; 6. studies on further absorbers like dust needed; 7. measurements at undisturbed reference sites needed) are not related to this study and were actually known before. Only the third recommendation is directly linked to the performed experiments.

The Referee is correct in that some of these recommendations were not of relevance to this study. In the revised manuscript some of the recommendations were omitted and the newly added ones were sharpened to more accurately reflect this study.

Change in the manuscript:

Recommendations 4-7 in the conclusions (section 4) were removed and two newly sharpened recommendations were added.

P. 1232: The authors claim in the introduction that the “focus is on the effects of the artificial impurities right after deposition, without further snow-soot interaction processes.” However, the majority of chapter 3 deals with trends in the BC (or EC) concentrations during the different experiments, trends in the snow albedo, and changes in the physical properties of the snowpack. These discussions are backed by table 2 showing BC concentrations during the SoS13 experiment on three different days and figures 2 and 3 showing snow profiles also during the SoS11 and 13 experiments and time series of albedo and meteorological measurements for SoS11 and 13 from the start of the experiment until the melting of the snow. I actually find the development after the initiation of the experiments the most interesting part and it makes sense to focus on these trends, but this is in direct contrast to the statement in the introduction.

Thanks for pointing this out. This statement in the introduction is misleading considering the results we continue to discuss in the paper. As mentioned earlier, the focus on the paper has been changed to the albedo changes caused by the soot deposition, which also include a temporal discussion.

Change in the manuscript:

The statement from the introduction has been removed for more clarity, and the focus of the paper is now more accurately reflected in the introduction.

Page 1232: Even in the Himalayas BC in snow concentrations above 100 ppb are rather exceptional. No references are given for the claim that in the Himalayas and the European Alps can be higher than 100 ppb.

The concentration of BC in snow and ice depend on several factors. These include the method of determining the BC in the snow, time of the year the sample is gathered, elevation from which the sample is taken (applicable primarily in mountainous regions), post-depositional processes taking place, and local and or regional emission sources.

In a recent paper by Kaspari et al. (2014), they report m BC (measured BC; which is an underestimate of the real BC concentration due to melting of the snow sample before analysis and the measurement technique used to determine BC) an average concentration of 180 $\mu\text{g/L}$ at Mera La (5400 masl), Himalayan Nepal. This was at their lower sampling site, out of three sampling sites on the glacier, where concentrations are expected to be higher. A max BC concentration of 318 $\mu\text{g/L}$ was measured at the Mera High Camp (5800 masl), indicating that such concentrations do occur in Himalayan snow even at higher elevations. Xu et al. (2012) also reported BC concentrations in the range of 11 to 3000 ng g^{-1} on a glacier in the Tien Shan Mountains, on the Tibetan Plateau. The extreme values were encountered in the surface layer snow during summer (during which post depositional processes such as melting as sublimation amplify the BC concentrations at the surface). Similarly, Qu et al. (2014) present BC concentrations above 100 ppb from Zhadang glacier, the Tibetan plateau, China. Average concentrations of 472.6, 334.4, and 142.9 ppb are presented from three locations of the glacier in July of 2012. Yet, other references from the Himalaya's report BC concentrations below 100 ppb, see e.g. Ming et al. (2013). Overall, the Himalaya's is a very large area where additional measurements of BC in the snow are crucial to better decipher BC concentrations, as the complete picture is quite complex.

For the European Alps, two references (Lavanchy et al. 1999 and Fily et al. 1997) were found with reported soot concentrations over 100 ppb.

Change in the manuscript:

The references from the Himalaya's and the European Alps with BC concentrations about 100 ppb discussed above have been added to the paper in the introduction.

Page 1234: The SoS2012 experiments are described, but the only results used further seem to be the characterization of the BC particles (size distribution, SP2 measurements) while still in the gas phase in the cylindrical chamber. (By the way: How reliable are the SP2 measurements made inside the chamber? Any effects due to the walls? No further descriptions of the measurements are given, nor of any details how the results were derived.) However, in the SoS2013 experiment the blowing system to transfer the soot into the air was modified impacting the size distribution of the particles. This leads to a couple of questions: How useful are the SoS2012 measurements for the SoS2013 experiments? The authors claim that changes were either small or only concerned the largest particles (page 1235). Was this tested? If yes, how? If they are not comparable, the description of the SoS2012 experiments may as well be deleted.

The changes to the blowing system between SoS2012 and SoS2013, and the subsequent changes to the soot deposited onto the snow surface are not well known. Our best estimate is that the larger sized-particles are the ones mostly affected, but no tests were performed to verify this. Such test would have been needed for us to make such claims.

Change in the manuscript:

In the revised manuscript we have deleted the soot characterization from the SoS2012 experiments (section 2.3). Since no results or data from SoS2012 will be used we have decided to only mention that it was done, but that no results are presented in this paper. This has not negatively affected the manuscript.

Pages 1237/8: At which height were the albedo measurements made? What is the field of view of the downward looking instruments? How do the fields of view correspond to the manipulated area? Any impact on the measurements due to the strong differences in BC inside and outside the manipulated area? All these details that may be important for the interpretation of the data are missing. In contrast, the second paragraph of chapter 2.4.2 describes spectral albedo measurements that are not used in the manuscript but have been (or will be) presented elsewhere. This paragraph could be deleted without any impact on the manuscript.

The field of view of the pyranometer is 2π steradians. This means that photons impinging on the receiving surface underneath the quartz dome may come from any solid angle within the whole 2π hemisphere below the downward looking sensor. However, when determining the measurement height, it was required that the potential specular component of the reflection should emanate from the deposited surface throughout most of the day, in practice at solar zenith angles <80 deg, when also the levels of both the downwelling and the upwelling irradiances are well above the detection limit and distinguishable from noise. On the basis of considerations accounting for both the position of the Sun and the dimensions of the deposition area, the measurement height was set to 30 cm.

Due to the large field of view of the pyranometer, we acknowledge that reflections from the surrounding non-deposited snow surface have an impact on the measurements of reflected global radiation above the deposited area. Indeed, we consider the deposited areas causing disturbances to the local albedo that we aim to derive through the measurements of upwelling and downwelling global irradiance. We expect the magnitude of the disturbance to depend on several factors like the concentration of the initial deposition, grain size and water content of the snow, migration of the soot into the deeper layers of the snow pack etc. Measurements over pure non-deposited snow represent the reference case, whereas measurements over deposited areas yield the cases of disturbance caused by deposition.

Exact calculation of the impact of the non-deposited area on the measurements performed over deposited area would require exact knowledge on the angular distribution of the upwelling radiation. Even though that knowledge is not available to us, we may roughly estimate the impact as follows: Let us assume a) ideal cosine response for the sensor; b) purely isotropic reflections from the surface; and c) that the deposition causes reduction in reflectivity that can in this idealized case be quantified as a fraction c of the reflectivity of pure snow. Let us use the notations given in Fig 1. as follows: h is the measuring height, and R_d is the radius of the deposited area. Let us also choose the horizontal position of the sensor directly above the center of the deposited spot. The sensor receives the radiation reflected by an infinitesimal surface area element proportional to the sine of the angle $\pi - \alpha_d$ between the point of reflection and the normal to the sensor. Irradiance received by the sensor is therefore proportional to the following integral expression:

$$\int_{-\pi/2}^{-\alpha_d} \cos \alpha \, d\alpha + \int_{-\alpha_d}^0 c \cdot \cos \alpha \, d\alpha = 1 + (c - 1) \sin \alpha_d, \quad (1)$$

where

$$\sin \alpha_d = \frac{R_d}{\sqrt{R_d^2 + h^2}}. \quad (2)$$

The amount of reflected global radiation measured at height h above a deposited area with radius R_d is hence found to be proportional to factor

$$1 + (c - 1) \frac{R_d}{\sqrt{R_d^2 + h^2}}. \quad (3)$$

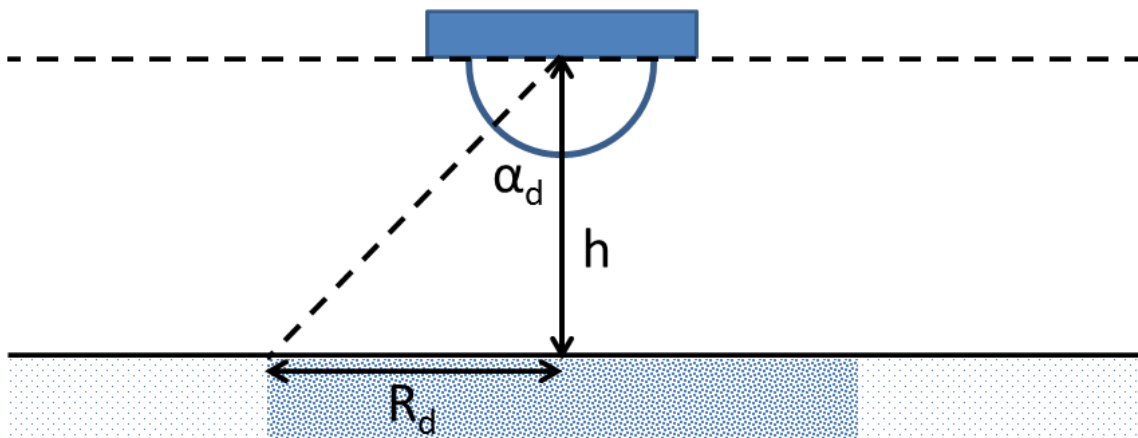


Fig. 1. Schematic presentation of the geometry of the experimental setup used for measurements of reflected global radiation.

The behavior of this factor may be examined by giving fixed values to c , R_d , and h . As an example, we choose $h = 0.3$ m, as this was the height used in the experiment, and $c = 0.625$, as the fraction of 0.5 (albedo assumed for deposited area) and 0.8 (albedo assumed for pure snow).

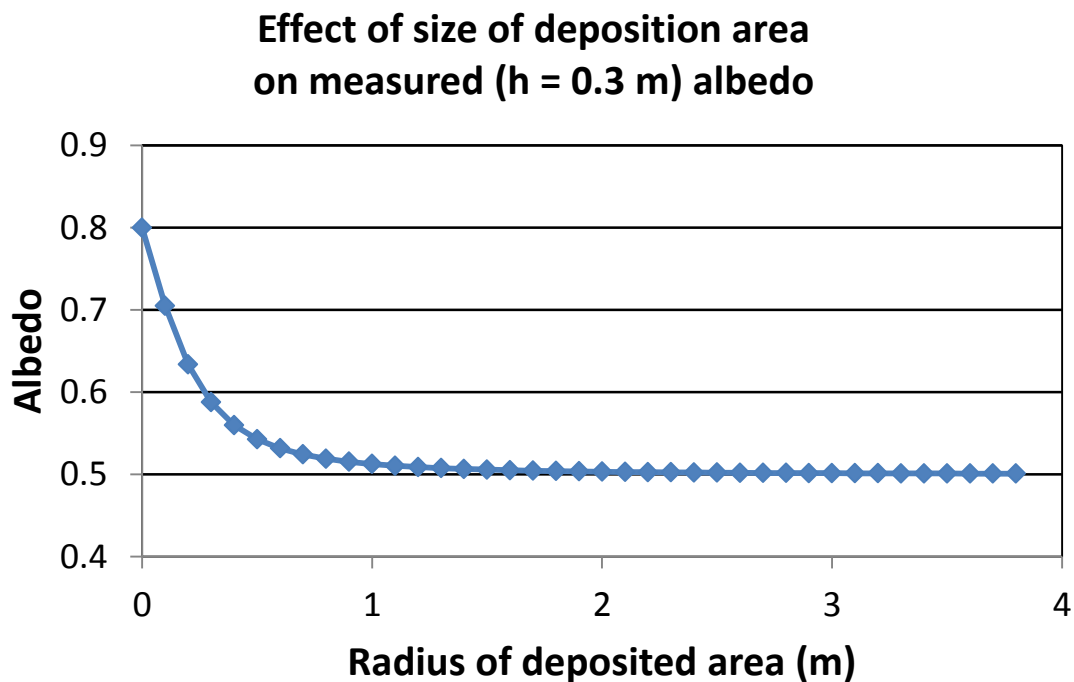


Fig. 2. Effect of the size of the deposited area on the albedo measured at the height of 0.3 m, assuming the albedo of pure snow to be 0.8 and albedo of deposited snow to be 0.5.

On the basis of this simplified approach, we may conclude that for the deposited areas of radius 2 m, for instance, the albedo measured at the height of 0.3 m is mainly dominated by the deposited area but indeed slightly affected by the albedo of the pure snow outside the deposition area. Obviously, the real anisotropic characteristics of the reflected radiation make the situation more complicated and may somewhat alter the dependence of the measured albedo on the radius of the deposited area.

Change in the manuscript:

The second paragraph of section 2.4.2 has been removed in the revised version on the manuscript as suggested by the referee. The contents of the paragraph has been expanded to cover the issues pointed out by the referee, including the measurement height, filed-of-view of the sensors, and the impact of the reflections outside the deposited area on the measured albedo. The BRF-measurements are now only briefly mentioned (as they are connected to the SoS-experiments) in the introduction and referenced to the paper by Peltoniemi et al. (2015) where they are further discussed.

Pages 1238/9: The authors report that snow temperatures, densities, SSA were performed. Why are the results are not shown in the snow profiles presented in Figure 2? For example, the SSA measurements were only used in the manuscript to derive an average optical radius for the SNICAR simulations. No further details concerning the SSA data are presented.

This mistake has been corrected and adjusted in the revised manuscript. Unfortunately the SSA measurements were only done before soot deposition in the one reference pit measured on 6 April 2013. This measurement of the SSA and the conversion to OEGD we estimated to best mimic the effective grain radius, needed as an input parameter for the SNICAR-model, which is why these SSA measurements were included in the manuscript.

Change in the manuscript:

In the revised manuscript the snow stratigraphy measurements are updated, now including the temperatures and densities, as well as the SSA when it was measured (see tables S1-S10). The snow stratigraphy profiles were reorganized altogether in the revised manuscript and include additional snow pits which were not presented in the earlier manuscript (see additional snow pit profiles in S3-S4, S8-S10).

Anonymous Referee #2

The work presented by Svensson et al. focus on the impact of black carbon (BC) in snow on the albedo and the snow melting. They artificially doped the snow with BC and measure the snow albedo, the BC content and the snow physical properties. The experiments were performed with a different setup each time and at three different location and during three different periods of the year. The authors concluded that in general albedo measurements and simulations using SNICAR agree. BC in snow has been studied since a few decades now but still our knowledge about its potential climate impact is poor. Of a particular interest is the melting season and the behavior of BC: would it stay at the surface or not? Does BC affect the physical properties of snow? This question is crucial and such experiment as the one conducted by Svensson et al is one of the first step in understanding such issue. Unfortunately, the presented experiments and the data analyses is very limited. In short, there is everything in that work: BC measurement, albedo measurement, snow physical measurement but there is never all in once and some aspects of the sampling protocol are very questionable. It is either some BC measurements, but the snow profile is made one week before, the snow physical data are not presented and there is no albedo measurement on the site used for following the BC concentration at the surface (Site 5 and 7). Even if the topic and the experiment is interesting and very few group are focusing on that, the results presented by the group do not bring new knowledge to the snow BC community and I can only recommend to refuse this paper for publication in TC.

Some specific comments below:

Page 1229, line 26: The authors wrote that they wish to focus on the effect of soot on snow properties but in the current paper, even if physical properties of snow such as density and snow SSA are claimed to be measured, there is no value presented at all.

Similar comments regarding the snow properties and results not presented were given by referee #1. This issue has been addressed in the revised manuscript by adding results and reorganizing the data.

Change in the manuscript:

The density and temperature have now been added to the new snow stratigraphy tables, which have been updated and reorganized altogether for more clarity in data presentation (see tables S1-S10). Further, an explanation to the SSA measurements has been provided. In the revised version of this manuscript it is also emphasized that the focus of this paper is on the effect of soot on the snow albedo, particularly the broad band albedo, in a natural setting brought by the nature of our outdoor experiments. The consequent melting of the snowpack and the effects of soot on the snow properties were also observed, however, not the focus of this study. This has now been changed in the introduction of the revised manuscript.

Page 1233: the three experiments have been run using different sites each time, with different snow depth and some most likely not optically semi-infinite for good albedo measurements and also at 3 different period of the year. This is of course not always easy to organize experiments but as all three experiments are in a way different and with different amount of incoming solar energy, this render the comparison event more difficult.

Direct comparisons between the different experiments are rather difficult and the referee hint at some of the difficulties. When conducting outdoor experiments these are factors that one is faced with compared to a laboratory study when the variables are more easily controlled. Still, the idea behind these experiments was to conduct them outdoors in ambient conditions to see the effects in this type of setting. For the obvious different meteorological conditions existing in northern versus southern Finland, the timing of the experiments had to

be different. In the revised version of the manuscript we have decided to remove the 2012 experiment (based on comments from referee#1). The results for 2011 and 2013 are therefore only considered, and direct comparisons between these two experiments are rather seldom in the paper.

Change in the manuscript:
No direct changes necessary.

Page 1234: line 10. Where are the images? At least one would have been interesting to see the type of soot you have been using compared to what is found in the Arctic.

A microscopic image of the soot deposited onto the snow is now provided in the revised manuscript.

Change in the manuscript:
Fig. 3 has been added to the manuscript and section 2.3.3 describing the microscopy technique and results have been added.

Page 1238: Albedo measurements. What was the height of the sensor? What were the conditions during measurements, meaning is there any visible differences between cloudy and clear sky conditions on albedo data? The second paragraph describing the BRF measurements is useless in that paper as the data are not used but if you have been measuring spectral albedo, it can be interesting to: 1/ compared with the broadband value; 2/ used for albedo calculations together with the snow SSA and the BC profiles but a multi-layer model should be used for that (not the online SNICAR version you used, see comment below)

The height of the sensor has been answered to referee #1, see pages 5-7 in this document for our response. In the revised manuscript cloudy versus clear sky conditions has been indicated and discussed. The BRF measurements have been taken out of the manuscript. The suggestions regarding the spectral albedo are interesting and would be a quite valuable addition to the literature. We have decided, however, to remove the spectral albedo description from the paper and are planning to present the results in another manuscript in preparation.

Change in the manuscript:
Clear sky conditions have been highlighted in the revised manuscript by the addition of figure 5 and discussed in section 3.2.3. The description of BRF and the spectral measurements have been omitted from section 2.

Page 1238: Snow physical properties. Where are all the data mentioned to be measured?

The revised version of our manuscript contains updated snow stratigraphy pits where all measured variables are presented.

Change in the manuscript:

The snow pit logs now contain all of the measured physical properties of snow from that event. Also, two additional snow pits sampled before soot deposition in SoS2013 are presented, as well as three snow pits from the melting period (17 April 2013). The snow stratigraphy pits are presented in the supplementary materials.

Page 1239-1240: The use of Snicar: you have been using a single value for the snow grain size and for the BC then as SNICAR is a single layer model. Why using that model if you have been measuring vertical profile of BC? You mention you have been using 270 or 750 microns for the snow surface optical grain radius. This is quite old snow considering the size of the grain, it gives a snow SSA lower than $15 \text{ m}^2 \text{ kg}^{-1}$, this is the range for metamorphosed snow or depth hoar. You also mention a density of 200 kg.m^{-3} , this is quite low and it does not fit very much to the size of the snow grain you have been using. However, the equation 1 you have been using is not for the optical grain radius, but for the diameter. The correct formula to get the snow optical radius from the SSA is $3/(\text{ice} \cdot \text{SSA})$. Maybe it explains something. . .

Thank you for pointing out our mistake with the optical grain radius. We have updated this error in the revised manuscript as well as used a different value to our density in the SNICAR. The idea was to use SNICAR as it is the most commonly used model in BC reduction of snow surface albedo. Measurements of the vertical profile of BC in snowpack were performed, however, these were not the focus of this manuscript (and are even less emphasized in the revised manuscript). We were more interested in the surface albedo which makes SNICAR sufficient for this purpose.

As an upper estimate for the grain size we used $750 \mu\text{m}$ in the manuscript, but do realize that this is quite large and probably unlikely as the effective grain radius that is to be used in SNICAR. This connects to converting the macro-photograph size (or visually estimated for that matter) to the effective grain radius used in SNICAR. In the revised manuscript we used $500 \mu\text{m}$ as the upper limit in the SNICAR model, and this number was based on the average macro-photograph size for the 1-9 cm depth interval measured in the same snow pit as where the OEGR was measured.

Change in the manuscript:

The SNICAR-model simulations have been rerun with a corrected grain radii, density, and height of the snowpack. The optical grain radius section in the text has also been corrected with optical diameter instead. The SNICAR model has also been run with $500 \mu\text{m}$ snow grain radius as a new upper estimate from grain size (instead of $750 \mu\text{m}$ in the older manuscript). Section 3.4 has been rewritten with regards to these changes in SNICAR.

Page 1241: how accurate is a measurement of 29 000 ppb of soot in snow using thermo-optical method? You probably only need a few milliliter of melted water to not saturate the instrument and as your deposition method is fairly heterogeneous in 2011, how this can represent the BC value at the snow surface?

It has been shown in previous studies that filters with a heavy aerosol loading make the split between OC and EC during analysis in the thermal-optical method less accurate (e.g. Cavalli et al., 2010). From this perspective the referee is correct that this measurement might not be truly accurate. During analysis, however, a smaller filter punch was used not to saturate the instrument and the thermogram from analysis demonstrates that it was not saturated. For this sample also only 51 mL of melted snow was used. Our estimate is that this sample is showing the right order of magnitude. Since the soot deposition was heterogeneous we used an average EC

concentration from 10 samples taken from the spot around where the pyranometer was set-up. The full range of these samples is now presented in the revised manuscript in section 3.1.

Change in the manuscript:

The addition of the full range EC samples from SoS2011 are now presented and discussed in section 3.1.

Page 1241: why there is no albedo measurement conducted on spot 5 and 7 while these spots are used to make the temporal evolution? Why don't you measure the albedo before deposition as well? Why don't you measure the albedo of the ground as well once the snow is melted and use the correct value as input in the model?

For the spots where albedo was measured we wanted to disturb as little as possible not to run the risk of spoiling or somehow influencing the albedo data. For this reason spots 5 and 7 were created to enable us to study the temporal evolution of the soot and the snow properties. The albedo was measured before soot deposition in spot 9b. It was after a few days of continuously measuring that this spot got contaminated. The albedo of the underlying ground was also measured since the devices measured until the snow was completely melted. In Fig. 4b this can be seen when the different spots reach their lower limit. Different albedos of the underlying ground were also tested in SNICAR but did not affect the overall albedo since the snowpack was thicker for these simulations.

Change in the manuscript:

No change needed.

Page 1242 and Table 2: Why does the sampling has been done in the 5 top cm and then the 7 top cm? How can you sum up the BC values together in the snowpack? Simply doubling the amount of measurements would then give you twice more BC then! In fact, you should have sampled the entire snow column (in several samples) and this is the only way to compare things and see if the BC is moving or not. Did you sample to the bottom? We do not know how thick the snow is on that date but I guess you are not at the bottom at 25 cm depth since they were still roughly 50 cm on 10 April (Stratigraphy data).

The difference in the sampling depths (7 versus 5 cm) of the snow was because of different sampling techniques used at the different measurement events. The sampling of EC right after soot deposition was done in 7 cm intervals, while it was done in 5 cm intervals during the later stages (10 April 2013 and 17 April 2013). Direct comparisons between them are not done in the manuscript. Summing up the BC values was done in a wrong manner and this has been taken out in the revised manuscript. Additionally, the movement of the BC particles in the snowpack and its discussion has been removed (see also answers to related comments given by referee#1, page 3 in this document).

Change in the manuscript:

Snow pit profiles have been updated with snow depth, and the corresponding soot measurement.

Page 1243: albedo measurements. Why are you discussing the data from 2011 while there are no BC measurements presented in the paper? Why fig 3 comes before fig 2? If you visually observed that the particles

have been sinking into the snow, this should have been sampled and would have been interesting for your purpose. How can happen that the precipitation event of 14 April brings the albedo of the most contaminated site higher than all others site, even the reference one? About the 18-19 April, following your hypothesis, liquid water lowers the snow density or wash out some BC, resulting in an increase of the snow albedo. How do you explain that all the albedo data are increasing of about 0.05? If the BC is washed out, the site with the highest BC content should increase the most. Else, if the density is decreasing, even if you have smaller optical snow radius, the radiation penetrates deeper in a lower dense media, so this could counterbalance the effect of the decrease of snow grains radius; and if the radiation penetrates deeper in the snow, it can encounter as well more BC particles. . . Again, your hypothesis could have been verified using the density data, claimed to be measured but not presented. When you discuss snow albedo, the snow depth is never mentioned.

Revised EC measurements in the snow from 2011 are presented in the results section (3.1) and table 2. The albedo section from SoS2011 (3.2.1) has also been updated with some new sections for clarity in the revised manuscript. We did not observe anything strange with the order of figures 3 and 2. The sinking soot particles should have been measured, but that was not the case in this study. It was not expected that the process would be this fast. Our hypothesis about the April 14th event is that the most contaminated soot spot had obtained a rougher surface structure than the other spots. Roughness itself would cause a decrease in albedo, but the uneven surface structure would more easily attach new fresh snow and sustain the accumulated snow better against snow drift compared to the other spots (if there was any wind). With more fresh snow the albedo would rise higher than the other spots. As mentioned this is a hypothesis and no measurements from the experiments are able to confirm this.

The precipitation around 18 April was rain and it was the most intense throughout the entire melting period. It is quite likely that some BC was washed out of the surface layer, however, no measurements are able to confirm this. Another possible reason for the rise in all of the spots is that the pyranometers were lowered by 20 cm to a new measurement height. The height of the sensors were meant to be lowered with melting snow to maintain this 30 cm above the snow surface. Around this time when there was such intensive melting maintaining these 30 cm were not carried out.

Change in the manuscript:

The EC measurements from 2011 have been rewritten in section 3.1. Section 3.2.1 has also been modified for more clarity. For the two events in SoS2013 (14 April and 18 April) our hypotheses have been further explained in the text (section 3.2.2).

Page 1245: snow physical properties. Finally you present a value of density, in the range of 340-400 kg.m⁻³ for the two snowpits made, so why have you been using 200 kg.m⁻³ in the modelling work? Where are all the snow physical data? Fig 2C is 2D. Figs 2D, E, F and G present the thickness of the layer and not the depth!

The two snow pits with a density range of 340-400 kg m⁻³ were from SoS2011 sampled one month after soot deposition, i.e. this contained older metamorphosed snow. The modelling work was done during the earlier stages of the SoS2013 experiment (soon after the soot had been applied to the snow) and so the measured variables from that stage were used as input to the model. In the case of the density it has been updated to 228 kg m⁻³. The snow physical data has been updated and is presented in tables S1-S10 (see also response to

the same comments above p. 10-11). The depth in Fig 2C-2G has been corrected to depth layers in the new snow pit stratigraphy.

Change in the manuscript:

In the revised version of the manuscript updated snow pit stratigraphy tables are presented (tables S1-S10).

Page 1247-1248: Using this model only allow you a single layering! Using your parameterization of equation 3 gives an albedo of 0.86 with 100 ppb of BC and 0.92 for 50 ppb of soot, so there is also a lower limit in your work. How did you end up with a linear regression fit while all precedent work, and the SNICAR model as well, showed a non-linear relation between BC and snow albedo? I also do not think that 7 points are sufficient for such sensitive topic as BC on snow.

The linear regression fit was also addressed by referee #1 (p. 3 in this document). A different approach to the fit has been done, resulting in a non-linear fit in the revised manuscript. We agree that seven points are rather limited for this topic, however, we wanted to make an attempt to put our data in a broader perspective.

Change in the manuscript:

A new non-linear fit is shown in Fig. 7, and further discussed in section 3.4.

Referee # 3

The manuscript by Svensson et al. is about the effect soot has on a natural snowpack. This manuscript is adding to the still limited literature of validating the effect soot in snow has on the snow pack physical and optical properties. Svensson and his group have performed experiments in the nature where they have added soot particles to the natural snow pack during three different times at three different locations in Finland. Each of the experiments has been different, both regarding the set-up, the soot applied and the measurements performed. After the initial soot deposition, the group has measured snow albedo, soot concentrations and snow physical properties at specific intervals.

Despite soot on snow has been on the agenda, both politically and scientifically for almost two decades, we still have limited information on the evolution of the snow pack after soot deposition events. Almost all of the existing work is based on models, and little work exists to validate the models with real measurements, with a few exceptions, including this manuscript. Despite this, the manuscript has some major drawbacks connected to it, regarding the available measurements for answering these questions, but mainly due to the performed data analysis. It will require a major revision to prepare this manuscript for publication in TC, but if the authors did the job, I think this could be an interesting addition to the literature.

General comments to the authors

- The value with this experiment is all the measured quantities. You should provide an overview of the three experiments, with photographs, timeseries or tables (as most suitable) of the available data. Table 1-2 is a start for BC concentrations for SoS2013, and Fig. 3 for albedo and met data for SoS2011 and SoS2013. By providing the measurements in a more consistent form, other scientists more easily get an overview on what's available and can be used for other purposes as well.

This is a valuable point on the manuscript and our experiments. An attempt to gain more structure and clarity as to what was done in the experiments has been made through the addition of table 1 (in the revised manuscript), where the overall characteristics of the experiments are given. From this table it is further referenced to where relevant data can be found (figure or table) in the revised manuscript. The snow stratigraphy measurements have also been changed and restructured to obtain more transparency in the paper.

Change in the manuscript:

Table 1 from the older manuscript was replaced by a new table describing the overall characteristics of the experiments. Snow stratigraphy figures in the old manuscript have been updated to tables in the revised version, as well as the addition of some snow stratigraphy measurements which were excluded in the old manuscript version.

- The soot concentrations are used in a wrong manner, eg. by adding concentrations without taking into account the depth or snow amounts these concentrations were calculated for. This needs to be corrected. If you have the concentrations for all layers, please make up bulk concentrations/amounts. If not the concentrations have to be used with cautions as they are of course dependent on the amount of snowfall.

With the comments given by referee #1 and referee #2 related to this topic, we have changed our use of the EC concentrations and feel that we have addressed this issue. See page 3 and 12 in this document our approach to this.

Change in the manuscript:

Table 2 was removed. New snow stratigraphy tables have been constructed and include corresponding EC amounts (tables S1-S10).

- It would be useful to have some words on the bias and uncertainties connected with the albedo measurements, in a similar manner as is done with the BC measurements (Sec 2.4.1). In addition you need to describe the field of view and the height of the sensor, hence how large is the area the optical sensor sees. Then you need a description on the homogeneity of the deposited soot for such a spatial scale.

We fully agree with the reviewer on the usefulness of an uncertainty estimate. In our case, where we study albedo as a function of two measured variables, the downwelling and the upwelling horizontal global irradiance, the work has to be carried out in two steps. In the first step, the uncertainties of the irradiances measured by the pyranometers has to be estimated. In the second step, it has to be estimated how the uncertainties in the irradiances propagate to the albedo values.

Prior publications on the topic indicate that there would be in principle two different approaches that could be adopted when estimating the uncertainty of the pyranometer measurements. The estimate could be based on general estimates derived as typical uncertainties for these particular instruments. Alternatively, a detailed analysis on all the potential sources of uncertainty, specific to the instrument and to the experimental setup as a whole, could be made, resulting in a comprehensive uncertainty budget for the irradiance measurements.

The manufacturer has made an investigation on the components contributing to the uncertainty budget and derived an achievable uncertainty of 3% for values of hourly integrated radiant exposure (Kipp & Zonen 2000, 2014). According to Kratzenberg et al. (2006), this is the representation of the expanded uncertainty commonly used by manufacturers of instruments of this type. This could well represent an estimate for the uncertainties encountered in our measurements as well.

However, it has been discovered by, e.g., Myers et al. (2004) that uncertainty in broadband solar radiometric instrumentation in general range from 3% up to 5%. Most notably, the uncertainty has been found to depend on whether a solar zenith angle specific responsivity or a constant responsivity is used. For our study, where we use the constant responsivity provided by the manufacturer, this would indicate that instead of the lower limit (3%) of the range of the uncertainty reported by Myers et al., we should rather use the upper limit (5%).

Given the rationale explained above, using the overall uncertainty of 5% for our pyranometer measurements could be perhaps justified in our study. However, we decided to engage ourselves into making a detailed analysis on the sources of all possible sources of uncertainties and compiled an uncertainty budget for our pyranometer measurements by following the guidelines given by the ISO GUM (JCGM 2008), and the specifications provided by the ISO 9060 standard. In practice, we adopted the same kind of approach and procedure as reported by Kratzenberg et al.

The analysis was based on the calibration certificates and the general information on the instrument provided by the manufacturer of the pyranometers Kipp & Zonen Inc (Kipp & Zonen 2000, 2014) . In addition to the

uncertainty of the calibration, the analysis accounted for the uncertainties originating from the following factors: Drift over time, directional response, offset originating from thermal radiation, offset originating from temperature changes, temperature dependence of the sensitivity, non-linearity, spectral response, tilt response, long-term drift of the measuring system, and errors in AD converter of the measuring unit. The budget was compiled on the basis of gross values of irradiance using the reference irradiance value of 500 W/m² since this was the irradiance used in the calibration of the instruments.

The CMP6 pyranometers used in SoS2013 campaign had been calibrated by the manufacturer Kipp & Zonen Inc. against their working standard pyranometers that are calibrated annually at the World Radiation Centre in Davos, Switzerland. The scale of the pyranometers is therefore traceable to the World Radiometric Reference (WRR). All the CMP6 sensors were new. The three CM11 pyranometers used in SoS2011 campaign were calibrated outdoors by the Finnish Meteorological Institute in 14th Jun and 3rd Jul 2012 against a reference standard instrument and following the guidelines given by ISO 9847:1992(E). The reference instrument was calibrated by WRR. Therefore, also the scale of the CM11 sensors used in SoS2011 campaign is traceable to WRR. The sensors were not used between the recalibration and the campaign. The calibration factors obtained in the 2012 recalibration were used in processing the measurements.

The calibration of CMP6 sensors performed by the manufacturer employs indoor calibration procedure based on a side-by-side comparison with a reference pyranometer under an artificial sun fed by an AC voltage stabiliser. According to the manufacturer, the overall (expanded, with coverage factor k=2) uncertainty of calibration is typically +-3% for the pyranometer of type CMP6 (Clive Lee, personal communication). The calibration certificates reveal that for our CMP6 sensors, the calibration uncertainty is +-2.4% (Kipp & Zonen 2011). The uncertainty of the calibration of CM11 sensors is typically +-1.5% (Clive Lee, Kipp & Zonen, personal communication). For the outdoor calibrations performed by the Finnish Meteorological Institute, it was conservatively estimated that the maximum uncertainty would be double of that, i.e., +-3% (Antti Aarva, Finnish Meteorological Institute, personal communication).

The most significant factor in the uncertainty budget of the CM11 pyranometers turned out to be the calibration. For the CMP6 sensors, the temperature sensitivity was found to be the most important contributor to the overall uncertainty.

On the basis of the detailed analysis performed, we estimate the expanded overall uncertainty (on confidence level 95%, two-tailed Student's test) of our CM11/CMP6 pyranometers to be **2.8% / (6.0-6.1)%**. These uncertainties indeed exceed the general estimate ended up by Myers et al. The uncertainties of the new CMP6 sensors seems unexpectedly high. However, an explanation to this is given by the fact that the CMP6 sensors belong to a lower category than the CM11 instruments. In ISO 9060 classification, CMP6 sensors belong to the group of "First Class" instruments, whereas CM11 sensors belong to the category of "Secondary Standard" instruments.

In the next step, we make use of the derived uncertainty estimates for the irradiance measurements to derive an uncertainty estimate for the albedo values we wish to study. We make the following notations:

I_0 = downwelling irradiance (measured by the pyranometer looking upwards)

I_1 = upwelling irradiance (measured by the pyranometer looking downwards)

A = albedo

The albedo is derived as the ratio of the upwelling and the downwelling irradiance. Hence,

$$A = \frac{I_1}{I_0}. \quad (1)$$

By using the error propagation equation (Bevington & Robinson, 1992), we get an approximation for the standard deviation in A :

$$\sigma_A^2 \approx \sigma_{I_1}^2 \left(\frac{\partial A}{\partial I_1} \right)^2 + \sigma_{I_0}^2 \left(\frac{\partial A}{\partial I_0} \right)^2 = \sigma_{I_1}^2 \left(\frac{1}{I_0} \right)^2 + \sigma_{I_0}^2 \left(\frac{I_1}{I_0^2} \right)^2. \quad (2)$$

Here, σ_{I_0} is the standard deviation in I_0 , and σ_{I_1} is the standard deviation in I_1 . As estimates for the standard deviations, we use the expanded standard uncertainties derived in the first step of the estimation process where we compiled the uncertainty budgets for the pyranometers. In other words, for 2011 measurements carried out by CM11 sensors, we use an estimate $\sigma_I = 0.028$, and for 2013 measurements having employed CMP6 sensors we use estimates $\sigma_I = 0.060$ - 0.061 .

Referee#1 also addressed the question on the field of view of the sensors and the effect of the non-deposited area around the deposited spot on the measured albedo. This has been discussed above (please see pages 5-7 in this reply).

Change in the manuscript:

We have added a paragraph dealing with the characteristics and uncertainties in section 2.4.2. The procedure used in compilation of the uncertainty budgets is described, and the resulting total expanded (two-sigma) uncertainties for the CM11 and CMP6 instruments are given. The field of view of the sensors is given and the effect of the FOV on the measurements of the reflected global radiation is discussed as well. Furthermore, the measuring height of the sensor and the criteria for determining the height are given. In the same context, the directional errors related to the deviations from an ideal cosine response are pointed out. In section 3.2.1 it is now pointed out that restricting the analysis to the noontime average values effectively removes potential effects of deviations from an ideal cosine response on the time series under study.

-Why compare to the single layer model SNICAR when you have measurements from several snow layers, and you are interested in how the BC percolates through the snow pack? There are other options available.

The idea was to compare the measured albedo of the surface snow with the commonly used SNICAR and so a single layer model would be sufficient. With the changes made to the revised manuscript, resulting in a stronger emphasis on the surface albedo and an even less focus on the BC movement within the snowpack, this model is the most suitable for our purpose.

Change in the manuscript:

No change needed.

- Why are the spectral albedos not used in this paper? Since the BC affects the spectral albedo mostly at the shorter wavelengths, investigating a range of wavelengths would make it easier to separate the effect soot has on the snow albedo compared to snow grain size or solar zenith angle.

This good idea would enable us to further study this. We have decided to omit the spectral data in the paper are a preparing another manuscript with the spectral data.

Change in the manuscript:
No change needed.

-It is extremely important to have a record on the snow physical and optical properties of both the reference site and the experiment site before the soot was deposited on the snow due, to the snow heterogeneity. Please discuss more about this, and compare the sites where you have this. For ex. page 1245, line 15: Where these sites compared and studied in detail prior to the sot deposition?

The sites mentioned on page 1245, line 15, were from SoS2011 (reference site and site affected by soot) and were not studied nor compared in detail before the commencement of the experiment.

Snow heterogeneity is of particular concern, especially when conducting experiments outside (compared to laboratory when homogeneous snow can more easily be constructed), and particularly when conducting experiments of this nature when you want to observe the effects on the physical properties of the snow. In SoS2013 the spatial heterogeneity of the snow was observed before the soot deposition through the multiple snow pits sampled prior to soot deposition. We have now included two more snow pits conducted from before soot deposition which were excluded in the previous version of the manuscript (see tables S3 and S4 in revised manuscript). We do admit that we failed with these experiments to make all of necessary snow observations in order to make sound conclusions about soot's effect on the snow properties. We have therefore in the revised manuscript emphasized that the focus on the paper was on albedo changes, and that the effect on the snow properties are included but not strongly emphasized.

Change in the manuscript:
Addition of some snow pit measurements done before soot deposition (tables S3 and S4), and the additional discussion section in 3.3 concerning the heterogeneity of the snow before soot deposition.

-Can you please elaborate how you dealt with the inhomogeneity of the soot in the 2011 experiment? My experience is that even for natural deposited BC we see large spatial variability, 30-50% within meter-scale.

Multiple surface snow samples (n=10) were collected to measure the heterogeneity in the sooted area for SoS2011. The average was 20885.7 ng g⁻¹ (standard deviation of 5198.5) and the minimum and maximum was 11072.5 ng g⁻¹ and 32865.4 ng g⁻¹, respectively. It was in this area where the pyranometer was set-up to measure the albedo. The clean reference area, which was not affected by the deposited soot had an average EC concentration of 79.8 ng g⁻¹, with a standard deviation of 31.0 (n=6). The maximum and minimum for this area was 135.1 and 43.6, respectively.

Change in the manuscript:

These values has been added (in table 2) and clarified in the revised manuscript (in section 3.1).

Some more specific comments

- Sec 2.3: What else did the soot consist of except BC?

This section was removed in the revised manuscript, but a new section on the electron microscopy work is presented in 2.3.2 to provide the read with an idea about the soot which we used.

- Sec. 2.4.3: How does the reported snow grain size compare with the optical grain size?

This is an interesting question that cannot be discussed in an elaborate manner in this manuscript due to the limited measurements. Measurements of the optical grain size and simultaneous traditional snow grain size were only conducted in one snow pit (reference snow pit from 6 April 2013 in SoS2013; see table S5a-b in revised manuscript). In the snow pit where it was compared the macro-photographed reported snow grain size generally overestimates by a factor of two compared to the optical grain size. This implies that overestimates (or underestimates, depending on which chosen) could be done when performing modeling work without carefully assessing the input variables for grain size.

- Page 1244, line 15 and about: It would have been useful to see the actual measurements from this event, not only the qualitative description.

Unfortunately we did not perform any kind of measurements of this event. We can only agree with this statement that it would have been very useful to present some quantitative characteristics of the event. No change in the manuscript needed.

- The literature clearly shows a non-linear relation between albedo and BC in snow. Why do you use a linear relation in Fig. 4?

The fitting has been addressed by all of the referees. It has now been adjusted in the revised manuscript.

Change in the manuscript:

A new non-linear fit is show in Fig. 7, and further discussed in section 3.4.

- Page 1238: There is a full paragraph (line 10-20) explaining measurements that are not used.

The BRF measurements description has been taken out in the revised manuscript.

- Page 1234: Line 18: How was this snow sample collected? Only surface sample or bulk sample? Please elaborate, and discuss this against the stated sentences. The new snowfall should not exclude this experiment from the analysis, IF the bulk (total depth) samples were collected.

These were snow samples from SoS2012, in which bulk samples were collected. Due to the other comments regarding the SoS2012 experiment, we have decided to remove any text related to the 2012 experiment, thus no discussion was added to the manuscript.

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