Author's reply to referee comments given on Svensson et al. "Contribution of dust and elemental carbon to the reduction of snow albedo in the Indian Himalaya and the Finnish Arctic."

Here we present point-by-point responses to the referee's comments. The referee's comments are presented in plain text in this document, while our reply is indicated in italics.

The consequent changes made according to the referee comments have improved the revised manuscript. Deletions in the manuscript have been marked by ~~strikethrough~~, whereas additional text are indicated by red color.

On behalf of my coauthors, yours sincerely,

Jonas Svensson

Anonymous Referee #1

The paper is focused on a significant and compelling issue: the attribution of light absorption in snow between primarily natural sources (mineral dust), and sources strongly influenced by anthropogenic activities (EC, especially in India). There are several main points made in the paper:

1) Mineral dust is perhaps the dominant light absorbing impurity in Himalayan snow

2) MAC of EC in snow is lower than for laboratory EC

3) MAC of EC decreases with increasing snow depth.

Although I imagine that points 1 and 3 have a good chance of being correct, all three of these conclusions suffer from some lack of support in the paper. I have substantial concerns about some of the assumptions and interpretations made in the paper, and question the conclusions that can be drawn from it.

My concerns are focused on the following issues:

1) The EC measurement of with the TOM method may be significantly affected by carbonates in the snow.

The authors cite Cavelli et al., 2010 to suggest that any contributions would show up as OC in the analysis, but this is not supported. First, Cavelli et al deals with atmospheric aerosol analysis in which carbonate "is generally <5% [of the aerosol]". However in snow, and especially in the Himalaya, radically more carbonate can be present. For example see: Di Mauro, B., F. Fava, L. Ferrero, R. Garzonio, G. Baccolo, B. Delmonte, and R. Colombo (2015), Mineral dust impact on snow radiative properties in the European Alps combining ground, UAV, and satellite observations, J. Geophys. Res. Atmos., 120, 6080–6097, doi:10.1002/2015JD023287. Which indicates that mineral dust content in snow in the European alps varied from the cleanest conditions (3 ppm dust) to levels of 200 ppm. 3 ppm of dust represents a factor of 15 times more mass than the EC concentration estimates of Svensson et al in Figure 12 (which I estmate as 200 µg/liter = 0.2 ppm EC). Note, too, that Cavelli et al points out for calcite that it "will possibly be detected as EC", and pretreatment of ice core samples was a prerequisite for analysis in Ming, J., H. Cachier, C. Xiao, D. Qin, S. Kang, S. Hou, and J. Xu (2008), Black carbon record based on a shallow Himalayan ice core and its climatic implications, Atmos. Chem. Phys., 8(5), 1343–1352, doi:10.5194/acp-8-1343-2008. As yet, the uncertainty raised by this issue is unbounded. Obviously, high mass ratios of mineral dust to EC in the Himalaya that contribute significantly to the TOM EC measurement are a real possibility that must be seriously addressed; ideally the authors would test some filter punches of ambient Himalayan samples with and without the type of acid treatment used in Ming et al., to constrain this issue. Note that conclusion #2 above is only valid if the EC determination is accurate. Further, note that conclusion 1, is not weakened by the carbonate issue, but should be reframed to reflect the clear contributions of mineral dust to in-snow absorption in the Himalaya.

The issue of carbonates in samples (both air and snow) is complex and further work on the topic is needed. During OCEC-analysis with the Sunset instrument and using the EUSAAR_2 protocol (used here) carbonates show up as a fourth peak during the first phase when OC is detected, and so it will affect OC (as pointed out by Cavelli et al., 2010). In other protocols (as well as other instruments), such as IMPROVE there is a risk that carbonates will affect the EC detection stage. It therefore depends on which protocol and thermal-optical instrument used (IMPROVE is frequently associated with DRI-analyzer). Using fumigation is a possibility, as in Ming et al. (2008), but in our case in favor of conserving resources we have not made the tests with acids. Instead, we have inspected the thermograms from OCEC-analysis, by visually observing any fourth peak in the

first stage, and have not observed any fourth peak with significant contribution to thermogram area for our samples. Furthermore, a recent study by Zhang et al. (2017), actually made tests as outlined by the referee here. With a subset of their filter samples from Tibetan snow, Zhang and colleagues found little difference between samples measured with and without acidification (less than 20% discrepancy).

In our revised manuscript we have added this reference, and clarified the text that carbonates are assumed not to be present insignificant amounts in our samples.

2) The laboratory tests of EC and SiC do not strongly constrain uncertainties in evaluation of ambient snow samples. The authors present several tests to evaluate the performance of their three measurement types: OCEC-analyzer TOM determination EC concentration; PSAP measurement of loaded-filter light transmission before and after undergoing TOM analysis; and loaded-filter light transmission measured with the OCEC-analyzer. However, it is not clear how relevant these tests are to the conditions relevant to the ambient samples. Comparisons of optical depth measured with the PSAP and the OCEC were presented as if they indicate the validity of the EC determination from the optical measurement; in fact they only illustrate measurement precision since both instruments were quantifying the same thing (optical depth on the filter). Hence the statement "The good agreement between the two optically derived EC values suggests that much of the scatter seen in Figure 5 is due to the uncertainty in the analyzed content of EC using TOM (and FID)" is incorrect and misleading. The authors should re-analyze the optical/TOM EC comparison to attempt to ascertain the confounding influences of the SiC, even as an imperfect proxy for ambient mineral dust contaminants. Further, although the EC concentration was not known (page 10, line 5), the relative concentration was. Hence there should be better constraints on the influences of the SiC on the different determinations of EC loading; I expect this dimension of analysis also to be relevant to the ambient samples. Undercatch by the filter was mentioned, but not given sufficient attention to justify confidence in the dataset. This can be a major problem (close to 100% for some filters!), especially for BC which is largely in the <1 μm mode, and typically <0.5 μm; hence the statement in the paper "smaller particles normally contribute little to total particulate mass" is not relevant to BC and filter undercatch. Note that the linearity of optical depth with estimated EC loading does not constrain filter losses. At a minimum, the authors should test undercatch with their filters by refiltering post-filter liquid from one of their EC standards with a high-efficiency filter. See, for example, Schmitt et al., doi:10.5194/tc-9-331-2015â for a treatment of similar issues.

The laboratory experiments are highly relevant for the ambient conditions, although the exact properties of the mixture of aerosol on the ambient filters are not known. It is imperative for the interpretation of field measurements that we can demonstrate consistency in the method using laboratory conditions that are controlled. Every permutation of possible aerosol mixtures can of course not be tested and we chose a simple set up where BC and the mineral proxy have roughly the type of optical properties that can be expected in ambient samples. Without this step, interpretation of snow samples associated with many unknowns would be very difficult. It is true that a best guess MAC has to be used for the estimation of light absorbing dust mass concentration but the variability and thus accuracy of using a fixed MAC would be the focus of another study.

The statement that the "good agreement" in fig.5 between the two optical measurements indicate that most of the scatter between TOM and PSAP is related to where the OCEC instrument determines to place the split point between OC and EC, still holds. The reviewer is right in that we do not explicitly suggest the reason for this scatter. Undoubtedly, the fact that BC is mixed with a mineral could very well influence the results. This is also one of the underlying reasons for why the laboratory exercise presented in the study is so central. We clarify the statement by noting that the reason for the scatter could be related to the fact that BC is mixed with SiC.

Undercatchment by filters is a potential uncertainty with loss of small particles. Many studies (e.g. Lavanchy et al., 1999; Forsström et al., 2013; Lim et al., 2014; Torres et al., 2014; Kuchiki et al., 2015) have performed limited tests on this enduring issue, and an array of different results have been presented, with nearly 100% in-efficiency or efficiency to capture the particles on filters. Clearly, the issue is complex, depending on several factors and still remains unresolved, and the aim of this paper is not to solve it. For our work here we are confident that larger sized particles have been captured by the filter, whereas we are unsure how efficiently our filters have collected smaller sized (<100 nm) particles. In the revised manuscript the reviewers concern has been elaborated on further.

3) Conclusion 3 above is based on the results shown in Figure 13. In my opinion, the data are not statistically robust enough to support that conclusion, and the discussion in p. 23 lines 28-34. Due to these major issues, I believe that the abstract, discussion, and conclusions should be modified dramatically.

We are aware that this is based on one pit study only, which is not enough and a lot more work needs to be done to obtain better statistics. However, with our data, and the one snow pit, we demonstrate a possible tendency (and not trend with the data). In the revised manuscript our enthusiasm of the results have been softened, highlighting the need for more measurements and that our tendency is speculative.

Smaller comments:

Abstract: 1) Snow albedo is strongly dependent on many factors beyond LAI (eg snow morphology, snow depth, underlying surface properties); since albedo is not a focus here, I suggest that the first sentences be reformed.

It is true that snow albedo depends on many factors. Still, LAI have the potential to significantly affect the albedo, depending on the pre-existing circumstances. As referee #3 also suggested, this sentence is changed in the revised manuscript.

Introduction 1) Since the SP2 is not used here, including it at this depth in the introduction seems unnecessary.

Actually, we disagree with the reviewer. This paper have in it a significant technical aspect of understanding LAI in snow and it is therefore pertinent to describe other relevant methods with a sentence or two. We adhere to the referee's criticism by shortening this particular section in the revised manuscript.

2) The discussion of non-BC LAI (page 4, lines 21-35) should probably be introduced before discussing the measurement techniques.

That is a good point, thanks. This section has been moved in the revised manuscript, appearing before the measurement methods of LAI section.

Misc:

1) Page 6 line 1: What is meant by "protected" here?

A glass container with a lid was used. We changed the word "protected" to "enclosed" in the revised version.

2) How were the filters dried?

The filters were dried in ambient conditions inside petri dishes. This has been added to the text.

3) P. 7, line 6: I am surprised that it is difficult to gravimetrically determine the dust concentration, as dust is almost certainly the dominant mass source left on the filter (at least in the Himalaya, where the optical thickness was too large to be measured!).

It is true that dust can be the dominating mass of the impurities on the filters. From our tests weighing filters show that these filters are not suitable for this procedure. Too much variance (noise) is introduced to get a good signal to noise ration. Additionally, the quartz fiber filters used are very fragile, and so while filtering, mass can easily be lost.

4) More information about the SiC used is needed. Manufacturer? Color? Etc.

The manufacturer is Carborundum (company non-existent anymore), as already indicated and the color was light grey with hints of blue. In the revised we added the color.

5) Please specify in the figure axis labels the technique (e.g. "optical" or "TOM" used to extract the various quantifications. For example, on figure 3, the caption merely indicates the EC was from the OCEC instrument, but it was not immediately clear that this via TOM.

Thanks, it is now fixed.

6) Page 12 line 7: again, the atmospheric results are likely not relevant in the Himalaya. I think they can be cited only for the Arctic results.

Our intent was to state that our laboratory measurements which showed that SiC has 100 times less MAC than BC, agrees with previous reports from atmospheric measurements (on a general scale), where dust is less absorptive.

7) What is the author's estimate of the loading of mineral dust on the ambient filters? How does this compare to the range tested in the laboratory?

We can only compare them by their optical signal. We can say nothing about mass. To do the latter we need to know the optical properties of dust in India (or Finland for the other samples). Since we do not know, we have used the ratio of tau to say how much the mineral contribute. Our range in the laboratory probably cover the same range, but this is speculative.

8) Please provide estimates of statistical relevance of the fits (e.g. r^2 values).

In the graphs the r2 values are already presented.

9) I saw concentrations presented as µg/L as well as ng/g. Please harmonize to a single unit for the reader (obviously these are effectively the same. . .)

Good, this should be consistent and has now been changed.

10) Page 17: lines 12-18: this is a lot of speculation without much reason for it. Suffice it to say that your results, for a different area where high spatial variability can be expected, are higher than a previous estimate.

Yes, that is true. But still we wanted to highlight some of these potential aspects that affect the differences. In the revised manuscript the text is changed accordingly.

10b) Page 17 lines 19-26: please include broader arctic estimates of EC concentrations for context. E.g. Doherty et al., 2010 ACP, doi:10.5194/acp-10-11647-2010.

Since Doherty et al. use a different measurement method we think it is sufficient to put our obtained results into a greater context with the results from other studies using the same measurement techniques. The other cited works present results for the broader European Arctic, which was not noted previously in the manuscript. This is now emphasized in the revised manuscript.

11) I don't see the value of the comments of page 17 lines 27-33, or figure 8 at this point in the text. This appears merely a technical point about measurement precision (as discussed above).

We disagree with the referee. The text is needed here to provide further credibility in our approach to analyze the data in the manner that we have chosen to do, so we feel that they are necessary to be there.

12) Couldn't the differences discussed in page 18 line 4-17 also be due to EC overestimation for example due to carbonate impacts?

As pointed out above, carbonates will not influence the TOM EC estimate, but could cause overestimation of OC. We refer to the discussion above for this point (p. 2-3 in this document).

13) Schwarz et al. only presented a theoretical estimate of MAC, not any measurements. Hence this is only suggestive, but it is also possible that the filter selectively catches larger BC. Please consider removing figure 10, as this is quite peripheral.

That is correct about Schwarz and colleagues, and it the revised manuscript this has been changed. The set-up was the same for the laboratory filters and the ambient filters, therefore the bias towards larger sized BC particles would exist in both sets of filters.

14) Page 20, line 18: "modes" is clearer than "peaking fractions".

That is true, and it is changed in revised manuscript.

15) Please specify the wavelength of the optical depth measurement in the OCEC instrument. How does this compare to the 532 of the PSAP? In discussion of relative absorption of mineral dust and BC, it is important to continue to specify the wavelength range for which the discussion is relevant.

For the OCEC-analyzer the laser operates at 632 nm (stated on page 7 in manuscript). Most of the discussion is for the PSAP, which is at 526 nm. This has been added to the dust fraction section (3.2.2) in the revised manuscript.

16) Please be consistent in referring to the dust "absorption" fraction. For example, in the caption to figure 11, "dust fraction" could be misunderstood by an un-alert reader as a mass fraction.

Referee #3 commented on this as well. The caption has been changed.

17) Is it possible to add data points to figure 12 a, so that the spread in results will be obvious to the reader?

Yes, an updated figure including the data points is now included.

18) What is the explanation for the huge differences in EC via TOM and OPTICAL at the sub surface contaminant layer shown in figure 12B?

This comes back to the OCEC-analyzer and where it places its split point in EC TOM, which has been shown to be less accurate for filters with a higher aerosol loading (Cavalli et al., 2010).

Anonymous Referee #3

Svensson et al. use a combination of a thermal-optical detection method and a particle soot absorption photometer to determine elemental carbon mass, its mass absorption cross section, and the contribution of mineral dust to the optical thickness of light absorbing impurities in snow. They conducted a series of laboratory assays with chimney and standard soot, as well as two different types of mineral dust, and mixtures of the various components to test the instrumental set-up. Subsequently, the method was applied to ambient snow samples from the Finnish Arctic and two Indian glaciers. The main findings are: a) the MAC of EC in snow seems to be lower than that of the laboratory test soot; b) dust plays a larger role in light absorption on the Himalayan glaciers than in the Arctic; and c) the MAC of EC in snow seems to increase with deposition age.

Generally, the study follows a careful design and is well described. It addresses the very important challenge of how to quantify the albedo-reducing effect of elemental carbon and mineral dust and their combination in snow, and contributes to the understanding of uncertainties by highlighting the importance of accurately determining the EC MAC in snow. Nevertheless, the work has some shortcomings: It is not clear why the MACs of two very specific dust types are determined in the laboratory and discussed when this is meaningless for the ambient dust samples with unknown absorption characteristics. An explanation of why the MAC of EC in snow is variable is missing, even though this is featured as a main result. Also the observation of a potential trend in the EC MAC value with snow pit depth is not backed up sufficiently. There is only a small number of data points, only one snow pit is discussed and the statistical significance of the trend is not given. Further, the value of addressing Arctic and Himalayan snow samples together in this manuscript is not evident. Additionally, the manuscript language needs to be improved.

I recommend addressing the above and following comments before a publication can be considered.

General comments

The title implies a discussion on snow albedo reduction through EC and mineral dust. However, the manuscript does not provide actual values of snow albedo reduction but rather focuses on accurately determining the mass of EC in snow and the contributions to optical thickness of EC and mineral dust. I suggest changing the title accordingly.

Title has been changed.

p. 10, l. 16/22: Information on light absorbing constituents in SiC and stone crush are needed. The light absorption of mineral dust is strongly influenced by e.g., hematite, goethite etc. There is no point in determining a MAC of a "random" dust sample in the laboratory to apply it to the ambient samples with unknown contents of light absorbing constituents as the authors state themselves on p. 20, l. 13. It is not clear, why dust samples are tested in the laboratory that have no relevance for the ambient dust samples. Also, in Figure 7, the discrepancy between the gravimetrically determined SiC mass on filters and the estimated based on the MAC when values are > 7 g /m2 are just discarded without any further discussion. Is it possible, that the method does not work for high dust loadings? I suggest that the dust related aspects of the laboratory assays are drastically shortened to the information relevant for the ambient samples.

As previously stated to referee #1 the laboratory experiments are highly relevant for our work and are needed to demonstrate that our measurement method works in laboratory conditions before analyzing field samples. Please see our response to referee #1 on p. 3 where this is elaborated on. For higher dust loadings the method is less accurate, as expected, since there is a lower signal to noise ratio for the optical measurement.

l. 13 – 27: What about local dust sources? There are most likely bare rock or mountain walls from which mineral dust can be deposited on the glacier. In general dust sources and in particular local dust sources can be highly variable so that any kind of interpretation in terms of trends is difficult, especially if no information about the origin is available. If the authors had mineral dust size distributions at least a statement on potential role of local sources if very large particles are present could be made.

True, this sort of statement(s) could be made if we had such data, but that is not the case.

l. 28 – 34: The interpretation that the MAC is decreasing towards the top of the snow pit is not convincing and an explanation why this might be the case is missing completely. There are not enough data points to conclude a trend in the MAC. The profile rather shows that LAI deposition is highly variable. Towards the bottom of the snow pit the ratio is also lower and if the point at 10 cm depth were not as low as observed, probably no trend would be inferred. The authors introduced on p. 20 the hypothesis that potentially large loadings on filters play a role for the observed variability of the MAC (see comment further above). To test this and to develop a potential explanation why the MAC changes, I recommend plotting in Figure 13 the ratio of the optical and TOM EC versus the ratio of Dust/EC and OC/EC to check if there is a relation with the overall LAI content. Also, indicate in the figure which ratio is meant. In the conclusion, the potential reasons for the observation should be given as well.

We realize that this is based on one pit and that our data is not conclusive. Referee #1 also commented on this topic (p. 4 in this document), further details are presented there.

The authors recommend additional work to constrain the optical properties of EC in snow but do not say how this could be done. Some more precise ideas would be useful.

That is true. In the revised manuscript we have added a thought on this.

Specific comments

p. 2, l. 3: What do you mean with "impact on climate"? LAI deposited on snow do not directly impact the climate but rather the hydrological cycle. Only if seasonal snow and glacier retreat significantly, local climate will change and LAI are hardly the major cause for it.

We did not mean it the way the referee has interpreted the sentence. LAI will affect climate through its lowering of albedo and subsequent snowmelt. Nevertheless, the sentence is now changed in the abstract.

p. 3, l. 2-3: Specify which radiation budget you mean and through which mechanism snow melt can be enhanced?

Snow melt is enhanced via snow darkening that is induced by the deposition of LAI. This has been clarified in the revised text.

l. 6: "In this context" refers to mountainous glaciers. Why are mountain glaciers more important than seasonal snow or ice caps?

We do not mean to suggest that mountain glaciers are more important than seasonal snow or ice caps in any way, and the text "In this context" has been removed in the revised version.

l. 12-14: I suggest removing this sentence and focusing only on BC effects in snow. In addition, BC has health effects everywhere where a human being is exposed, not only in cities and where open cookstoves are used.

This is true, the sentence is now deleted.

l. 21: is Bond et al. 2007 really the most up to date reference?

Bond et al., 2013 is newer reference sufficient for this statement.

l. 34: more elaboration on the thermal-optical analysis is needed here.

At that stage in the manuscript we simply want to only introduce the method. An elaboration is provided in the methods section, which we feel is adequate.

p. 4, l. 3 – 8: The description of the SP2 can be shortened because this instrument does not play any role in the set-up of this work.

In the revised version it is has been shortened (as pointed out by the other referee, p. 4 of this document).

l. 10f: An explanation of why it is important to keep the samples frozen before analysis is needed.

Good point. Particle losses can be very significant if a sample is not kept frozen. In the revised manuscript we have added this.

l. 18f: The statement that small particles contribute little to particle mass in this context might be a bit misleading. More elaboration is needed. What is meant by small particles, what is the threshold that Lim et al. (2014) refer to? If it is < 100 nm the statement is ok, if it is < 300 for example, it would not be, because of the size distribution of BC. Also the last sentence does not take into account that the TOM method will also quantify OC, which can also absorb light and reduce snow albedo.

We are referring to fig. 3 of Lim et al. (2014), in which one can see how the filter efficiency increases with BC size. The issue of undercatch can be significant for quartz-filters and it is not resolved to-date. In the revised manuscript this has been modified.

p. 5, l. 21: atmospheric concentrations of what?

It refers to BC concentrations and other aerosol particles (particulate matter), which is stated earlier in the sentence, with both displaying seasonal patterns.

p. 6, l. 3f: A description of how the filters were dried is missing here and later on for the microwave heated samples as well.

As noted by the other referee also, this has now been modified in the revised manuscript.

Figure 1: The location of the glaciers and the valley is not really visible, the site symbols are too small and the lat/lon numbers in the middle of the plot are distracting. Also the location of the sampling sites in Finland are not visible at all. Think about including two maps that show the details for each region, while keeping the global map.

A new map has been made for the revised manuscript.

p. 7, l. 6: Why is it difficult to gravimetrically determine the dust load on filters? This is a standard method and the mass of dust very often is much higher than the mass of other LAI. Do you mean it is difficult in regions where little dust is present?

From our tests it is difficult to weigh the quartz-filters as there is significant variance. This issue was also commented on by referee #1, so please see our response on p. 5.

l. 33: It is unclear from the description what the original value of transmittance is.

It refers to the transmittance signal measured by the OCEC-analyzer right before the first step of analysis (OC detection). We have added the text "(measured before thermal sequence starts)" in the revised manuscript to clarify this.

p. 8, l. 5ff: Why are the authors confident that no acid treatment of the samples is necessary to eliminate carbonates? In ambient mineral dust calcium carbonate or other carbonates are a common constituent which might strongly affect the OC/EC analysis and not only the quantification of OC as you explain in l. 11f (split point).

Carbonates shows up as fourth peak during the first stage (OC-stage) of OCEC-analysis using the EUSAAR_2 protocol (Cavalli et al., 2010). This was not found for our samples after visually inspecting our thermograms from the analysis. See also comment by referee #1 and our response on page 2 for more information.

l. 8-12: Do the authors have any estimate of the loss of small particles at least for the laboratory experiments using the same filters? Since the laboratory pre-assays are meant to quality control the results of the ambient samples this is a crucial piece of information that should be obtained.

We do not have any estimate of the loss of small particles. This is a topic (undercatchment) that should be further explored, but not in this paper.

l. 18 – 21: From the description the methodology is not clear. Did the authors use duplicate samples, so 2 times original and 2 times heated, or two punches and one was heated while the other wasn't?

This is the general methodology of the PSAP and how it was applied for our purpose. It is further described on p. 10 in the manuscript. In the revised manuscript we have directed the reader that the methodology is further presented in section 2.3.

p. 9, l. 9f: What is the reason for this? Is the effect of the filter so overwhelming that the scattering effect of the actual sample is by far outweighed?

The reason is that the filters we are using do not have any known correction factors (compared to for example PSAP-filters that have existing correction factors in the literature). As we note in the previous sentence of the manuscript, we treat the enhancement factor as a constant for our samples, and so it is included in our effective MAC.

l. 20: It needs to be stated more clearly that the MAC values reported here cannot be compared to other studies.

This is now emphasized in the revised manuscript.

p. 10, l. 8: Information on the homogeneity of the chimney soot is missing. How comparable are samples? Why was chimney soot chosen, is it representative for soot deposited on snow in Finland and India?

Samples are comparable in the sense that when a larger amount of the stock solution was taken, the filter appeared darker (and had higher EC content). Chimney soot was chosen since it is easily accessible and is a

common pollutant in the study area, especially in the Himalayas. Moreover, if a mixture of chimney soot, vehicular soot and other soot, then what would be the appropriate proportions?

L. 35 f: What are twin samples, and what does "separate instruments" refer to? The meaning of the sentence is unclear.

The "separate instruments" refers to the OCEC-analyzer and the PSAP, and the twin samples refers to two identical samples. In the revised manuscript this has been clarified.

p. 11, l. 1: how many filters were tested and what was the result in numbers?

The number of filters was 7 with a difference less than 5%.

l. 19: What is divided by Cref?

It is the optical depth as noted later in the sentence.

Figure 4: How would the slope look like if the point with the highest loading of SiC were not considered? It drives the correlation result and how would results look like that are presented in Figure 7?. In the caption the wavelength information is missing.

Excluding the point with the highest SiC does not change the slope (it remains 0.23). The wavelength information is now included.

Figure 5: Add the 1:1 line.

In the revised manuscript this has been added to the figure.

p. 14, l. 8ff: The spread in the comparison of the two optical EC determination methods is not necessarily an indication for variability in the evolved carbon determination of EC. It is not clear what is meant with "uncertainty in the analyzed content of EC using TOM. . .". Which factors introduce uncertainty in this method? Which introduce uncertainty in the optical methods?

A similar comment was noted by referee #1. We are referring to that that most of the scatter between TOM and PSAP is related to where the OCEC instrument determines to place the split point between OC and EC. See further our comment to referee #1 on page 3.

p. 17, l. 11f: Concentrations are given in µg / L and ng/g, one consistent unit should be chosen.

Thanks, it should now be consistent in the revised manuscript.

Figure 8: Is the outlier at Sunderhunga taken into account for the linear regression? If so, what would the slope look like without? It would be even greater than 19 % and that is a relatively large deviation between the optical measurement of EC with TOM and the estimated EC based on the MAC and the PSAP data. What does this discrepancy mean for the validity of the methods?

The outlier from Sunderdhunga is included in the regression. Taking this data point out from the regression we obtain a slope of 1.31 (although we do not see the point of removing this one data point since it merely shows some scatter in the data). It could be argued that data points with an EC content higher than 0.03 g m^{-2} (ca. Tau=1) could be removed since at higher EC content there is a lower signal to noise ratio for the PSAP measurement. With this procedure we get an identical slope (1.18) as the original value in fig. 8 (1.19). Thus, removing data points does not affect the regression.

p. 18, l. 12: A reference and an explanation is missing.

This is the enhanced absorption effect of BC particles dependent on the mixing state. References have been added in the revised manuscript, and the sentence has been slightly modified for clarity.

p. 20, l. 9ff: The underlying assumption why EC is less absorbing in this case is not provided and not evident. Do the authors assume that the EC is embedded so thickly in OC that light does not penetrate to the EC? If the authors imply matrix effects of the sample, the question is how relevant this is to the ambient snow? On the filter the particle mass is concentrated and the packing of the particles on the filter is not representative for the packing in the snow.

We do not know the reason for the apparent reduction in absorptivity between our lab BC and ambient samples, this is why we offer different hypothesis for this difference. One of these is embedding BC particles in water-insoluble OC, the other is size effects.

It is most likely that the conditions on the filter differ from the conditions when the particles are naturally in the snow. However, we are not aware of any non-intrusive method to date that can determine LAI concentrations in snow and ice. In our study we never exploit our numbers into calculating radiative effects or climate impacts, but rather try to understand levels of LAI that can be present in snow and ice and advance our knowledge about possible processes related to this.

p. 21, l. 10: Do the authors mean the average EC concentration when saying "composite"? Be more clear.

Yes, that is what was meant, and is now changed in the revised manuscript.

p. 23, l. 3: How do the authors know that those are different seasons over various years and not melting and freezing cycles within one year? An explanation is missing in the text.

Concentration cycles as observed reflect seasons, while short-term events would produce more random distributions. In the previous sentence we offer an explanation.

l. 7: Water soluble constituents might percolate, this is not known because it hasn't been investigated. So there cannot be a statement that this is not the case.

The vertical LAI distributions indicate that not much mixing has taken place. We have changed the wording of the sentence and added a reference where this is discussed (Doherty et al., 2013).

Technical comments

p. 2, l. 3: write "with subsequent implications for snow melt. To more accurately quantify changes in snow albedo, . . ."

Changed.

l. 5-6: write ". . .from the Indian Himalaya and pared the results to snow samples. . ."

Changed.

l. 18: "dust deposition"

Changed.

l. 22: Do you mean deeper pits?

Yes, that was meant, and is now changed.

p. 3, l. 5: There is a newer AMAP report from 2015.

Yes, that is true, but that reference does not cover this topic.

l. 9: you should list rather fuel types than activities if you refer to carbon-based fuels.

We do not see the point in listing different fuel types. In the revised manuscript we added "in activities" to the sentence to clarify.

p. 4, l. 21: "can" instead of "may".

Changed.

l. 22: "microorganisms" instead of "microbiology"

Changed.

l. 33: "Other methods consist of using transmitting light. . ."

Changed.

p. 5, l. 1: "in the Indian"

Changed.

l. 4: tests

Changed.

l. 8: "2.1.1 The Indian Himalaya"

Changed.

l. 12: "valley-type glaciers in the Ganges basin. . . ."

Changed.

l. 14: delete "residing"

Changed.

l. 27: "Dust from local sources has. . ."

Changed.

l. 31: delete "designated"

Changed.

p. 6, l. 14: "sampled snow"

Changed.

l. 16: "(the first. . ."

Changed.

l. 17: ". . . where details of the area. . ."

Changed.

p. 7, l. 31: " for pyrolysis (darkening of the filter) occurring during the. . ."

Changed.

l. 34: delete "filters" before EC

Changed.

p. 8, l. 14: "uses"

Changed.

l. 32: write micrometer instead of millimeter

Changed.

p. 10, l. 2: Start the sentence with "A series of . . ."

Changed.

l. 5: replace "minute" by "small"

Changed.

p. 11, l. 15: "dependent on"

Changed.

l. 16: delete "thus many influences on it."

Changed.

l. 20: delete "somewhere"

Changed.

l. 21: "for our BC solution data"

Changed.

l. 22 "Bond et al. (2013) report. . ."

Changed.

Figure 3 Caption: "comparison of. . .", here and in several other captions. Also, sometimes OCEC-analyzer, TOM or Sunset analyzer is used. A more consistent use of the method name is needed.

Changed.

p. 12, l. 4: "Figure 4 shows results analogous to Figure 3. . ."

Changed.

p. 13, l. 9: "The data are scattered,. . . regression is within 17 % of the 1:1 line."

Changed.

p. 14, l. 7: "As observed, the EC amounts derived by two optically different methods show a consistent relation. . ."

Changed.

Figure 6 caption: "between the optical measurement of EC. . . on the substrate using PSAP data and the . . ."

Changed.

p. 15, l. 9: "Two slopes are presented, . . ."

Changed.

Figure 7 caption: "containing all data points"

Changed.

p. 16, l. 14: "with material and quantitative impurity. . ."

Changed.

p. 17, l. 2: "these surface samples contained LAI mostly originating from the post. . ."

Changed.

l. 3: "studies of BC"

Changed.

p. 17, l. 19: "For reference, the EC concentration in the surface. . ."

Changed.

l. 22ff: ". . . in Pallas might result from the fact that the majority of samples was taken later in the snow season. . . and EC has likely concentrated. . ."

Changed.

l. 25: "On a larger scale, Northern Europe and Arctic, the concentrations. . ."

Changed.

p. 18, l. 9: "a smaller absorption efficiency", replace "absorbing efficiency" by "absorption efficiency" here and in several other places in the text.

Changed.

l. 17: delete "originating"

Changed.

p. 19, l. 9: "demonstrates"

Changed.

l. 12: "for BC we use the same complex. . ."

Changed.

Figure 10: Explanations for the symbols in the equation are missing.

Changed.

p. 20, l. 4: "liquid"

Changed.

l. 7: "a scattering medium shows enhanced. . ."

Changed.

l. 8: "was" instead of "were"

Changed.

l. 13: exchange "applicable" by "possible"

Changed.

l. 18: "modes" instead of "fractions"

Changed.

l. 19: "with modes at 35 % and 65 %.

Changed.

l. 20: "by LAI other than BC. . ."

Changed.

p. 21, l. 3f: "reach as much as 56 %.. in the Tibetan. . . as a fraction of the optical depth of LAI on the filter,. . ."

Changed.

l. 5: delete "an" before albedo.

Changed.

Figure 11 caption: "Frequency of the occurrence of dust optical thickness fractions at the three sampling sites." Otherwise the mass fraction might be inferred.

Changed.

p. 23, l. 2: delete "evidently", replace "core" by "pit"

Changed.

l. 3: is "alternating" meant by "altering"?

Changed.

l. 8: replace "that" by "who"

Changed.

l. 13: replace "at best ca." by "potentially"

Changed.

l. 30: "decrease" instead of "be decreasing".

Changed.

p. 24, l. 14: "EC deposited on snow". The EC does not originate from the snow.

Changed.

l. 14 f: "Our finding of a MAC value of about half of . . . EC particles, can have implications for the snow. . ."

Changed.

References

Bond, et al. Bounding the role of black carbon in the climate system: A scientific assessment, J. Geophys. Res. Atmos., 188, 5380–5552, doi: 10.1002/jgrd.50171, 2013

Doherty, et al. Observed vertical redistribution of black carbon and other insoluble light-absorbing particles in melting snow, J. Geophys. Res., 118, 1–17, doi:10.1002/jgrd.50235, 2013

Forsström, et al. Elemental carbon measurements in European Arctic snow packs, J. Geophys. Res. Atmos., 118, 13614–13627, doi:10.1002/2013JD019886, 2013.

Kuchiki, et al. Elemental carbon, organic carbon, and dust concentrations in snow measured with thermal optical and gravimetric methods: Variations during the 2007–2013 winters at Sapporo, Japan, J. Geophys. Res. Atmos., 120, doi:10.1002/ 2014JD022144, 2015.

Lavanchy, et al. Historical record of carbonaceous particle concentrations from a European high-alpine glacier (Colle Gnifetti, Switzerland), J. Geophys. Res., 104, 21227–21236, doi:10.1029/1999JD900408, 1999

Lim, et al. Refractory black carbon mass concentrations in snow and ice: method evaluation and inter-comparison with elemental carbon measurement, Atmos. Meas. Tech., 7, 3307-3324, doi:10.5194/amt-7-3307-2014.

Torres, et al. Measuring Organic Carbon and Black Carbon in Rainwater: Evaluation of Methods, Aerosol Sci. Technol. 48: 239-250, 2014.

1 ~~Contribution of dust and elemental carbon to the reduction of snow albedo in the~~
2 ~~Indian Himalaya and the Finnish Arctic~~

3 **Light-absorption of dust and elemental carbon in snow from the Indian Himalaya and**
4 **the Finnish Arctic**

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1   **Abstract**

2   Light-absorbing impurities (LAI) deposited to snow have the potential to substantially affect the snow
3   radiation budget, with subsequent implications for snow melt. ~~have the potential to substantially affect~~
4   ~~snow albedo, with subsequent changes on snow melt and impact on climate~~. To more accurately
5   quantify the snow albedo, the contribution from different LAI needs to be assessed. Here we estimate
6   the main LAI components, elemental carbon (EC) (as a proxy for black carbon) and mineral dust in
7   snow from the Indian Himalaya and pared the results ~~compared it~~ to snow samples from Arctic Finland.
8   The impurities are collected onto quartz filters and are analyzed thermal-optically for EC, as well as
9   with an additional optical measurement to estimate the light-absorption of dust separately on the filters.
10 Laboratory tests were conducted using substrates containing soot and mineral particles specially
11 prepared to test the experimental setup. Analyzed ambient snow samples show EC concentrations that
12 are in the same range as presented by previous research, for each respective region. In terms of the mass
13 absorption cross section (MAC) our ambient EC had surprisingly about half of the MAC value
14 compared to our laboratory standard EC (chimney soot), suggesting a less light absorptive EC in the
15 snow, which has consequences for the snow albedo reduction caused by EC. In the Himalayan samples,
16 larger contributions by dust (in the range of 50 % or greater for the light absorption caused by the LAI)
17 highlighted the importance of dust acting as a light absorber in the snow. Moreover, EC concentrations
18 in the Indian samples, acquired from a 120 cm deep snow pit (covering possibly the last five years of
19 snow fall), suggest an increase in both EC and dust deposition, while at the same time ~~there~~ proposing
20 a tendency for a reduction in the MAC value with snow depth. This work emphasizes the complexity
21 in determining the snow albedo, showing that LAI concentrations alone might not be sufficient, but
22 additional transient effects on the light-absorbing properties of the EC need to be considered and studied
23 in the snow. Equally imperative is to confirm the spatial and temporal representativeness of these data
24 by comparing data from several and ~~longer~~ deeper pits explored at the same time.

1   1. **Introduction**

2   The deposition of light-absorbing impurities (LAI) in snow influences the radiation budget and can

3   cause enhanced melting via snow darkening (Warren and Wiscombe, 1980). This process affects

4   regions with seasonal snow cover, leading to an earlier snow retreat, which has major implications for

5   thawing and biogeochemical processes acting in the ground (AMAP, 2011). In mountainous areas with

6   glaciers, the impurities perturb glacier properties and the hydrological cycle (e.g. Xu et al., 2009). In

7   this context, The impact on snow reflectance (albedo) from black carbon (BC) aerosol particles is of

8   particular interest. Being one of the most effective light-absorbing aerosols, BC enters the atmosphere

9   by combustion of carbon-based fuels, in activities including forest fires and anthropogenic burning of

10   bio- and fossil fuels (Bond et al., 2013). Because of its negative effect on snow albedo, considerable

11   effort has been made to globally quantify BC in snow (e.g. Doherty et al., 2010; Ming et al., 2008;

12   Schmitt et al., 2015), as well as in ice cores (e.g. McConnell et al., 2007; Ruppel et al., 2014; Xu et al.,

13   2009). In urban areas and in households using open fires, BC particles are also known to have adverse

14   health effects, which make them interesting from a human health perspective as well (e.g. Shindell

15   2012).

16   The potential impact of LAI in snow and ice make the Himalaya a region of special interest. It contains

17   numerous glaciers which are in a general state of recession, although contrasting patterns have been

18   reported in different areas (e.g. Bolch et al., 2012; Kääb et al., 2012). Himalayan glaciers act as

19   freshwater sources for several major rivers in Asia, including Indus, Ganges, Brahmaputra, Mekong,

20   and Yangtze, thus having a vital part in millions of people's lives (e.g. Immerzeel et al., 2010). The

21   glaciers are especially susceptible to BC emissions, since India and China located in close proximity,

22   emit the most BC world-wide (Bond et al., 201307). A recent study by Ming et al. (2015) found a

23   decreasing trend in albedo during the period of 2000-2011 on Himalayan glaciers, and suggested rising

24   air temperatures and deposition of LAI to be responsible for the decrease. In light of the vast area of the

25   Himalayas, there is a lack of in-situ measurements of LAI on glaciers, which are crucial for modeling

26   work (Gertler et al., 2016). The lack of measurements is especially pronounced in the Indian Himalaya,

27   since previous measurements of LAI in Himalayan snow and ice have largely been confined to China

28   (e.g. Xu et al., 2006) and Nepal (e.g. Ginot et al., 2014; Kaspari et al., 2011; Kaspari et al., 2014; Ming

29   et al., 2008).

30   In addition to BC, other LAI can contribute significantly to the radiative balance of the cryosphere.

31   Recent research has identified mineral dust and microorganisms as having a more important role than

32   previously thought in the current decline in albedo of the Greenland Ice sheet and other parts of the

33   Arctic (e.g. Dumont et al. 2014, Lutz et al., 2016). Similarly, Kaspari et al. (2014) reported such high

34   dust concentrations in the snow of Himalayan Nepal that the contribution of dust in lowering the snow

35   albedo sometimes exceeded that of BC. The importance of dust has also been illustrated from other

1 regions, for example the Colorado Rockies, US, where dust causes a significantly earlier peak in runoff

2 (Painter et al. 2007). In the Arctic, Doherty et al. (2010) suggest that 30 to 50 % of sunlight absorbed

3 in the snowpack by impurities is due to non-BC constituents. Evidently, dust has an important role in

4 the cryospheric radiative balance. Differentiating between the different impurities in the snow is not

5 trivial, however, and requires more than one analytical technique (Doherty et al., 2016). Traditionally,

6 dust in snow has been quantified by gravimetrically measuring filters (e.g. Aoki et al., 2006; Painter et

7 al., 2012). Other methods consist of using transmitted light microscopy (Thevenon et al., 2009), a

8 microparticle counter to measure the insoluble dust (Ginot et al., 2014), or mass spectrometry (using

9 iron as a proxy for dust) (Kaspari et al., 2014).

10 At present, three primary methods are used to measure BC in snow and ice (see Qian et al., 2015, in

11 which they are extensively presented). Out of the three methods, two utilize filters to collect impurities

12 in a melted sample. The first filter method measures optically the spectrally resolved absorption by the

13 impurities using an integrating sphere integrating sandwich spectrophotometer (ISSW) (e.g. Doherty et

14 al., 2010; Grenfell et al., 2011). The second filter method is the thermal-optical analysis of filters (e.g.

15 Forsström et al., 2009; Hagler et al., 2007). The third, non-filter-based method, uses laser-induced

16 incandescence with a single particle soot photometer (SP2) (e.g. McConnell et al., 2007; Schwarz et al.,

17 2012).

18 Each measurement method has benefits and drawbacks. The SP2 is specific to refractory BC and is able

19 to provide estimates on the size of the BC particles. However, the SP2 has a size range limitation

20 (roughly 70–600 nm, depending on the instrument settings and nebulizer setup), which may result in

21 the underestimation of BC mass since particles in snow have been reported to be larger (Schwarz et al.,

22 2012; Schwarz et al., 2013). ~~Moreover, the SP2 technique needs to have the liquid particles aerosolized,~~

23 ~~which may lead to additional particle losses (Schwarz et al., 2012).~~ The use of filters, on the other hand,

24 can provide a practical logistics advantage for the collection of LAI in remote locations because it is

25 difficult to maintain the necessary frozen chain for the snow samples from the field to the laboratory

26 for analysis. Particulate losses can be very significant if a sample is not kept frozen, thus not providing

27 accurate results. Filtering of liquid samples can be conducted in the field, and the substrates are more

28 easily stored and transported to the laboratory. The ISSW method has the advantage that it measures

29 light-absorbing constituents on the filter indiscriminately. Thus, the ISSW method is not specific to BC,

30 and requires interpretation of the spectral response to determine the BC component. The thermal-optical

31 method (TOM) provides an actual measurement of elemental carbon (EC) that is instrumentally defined.

32 EC is assumed to be the dominant light-absorbing component of BC, and often EC and BC are used

33 interchangeably in literature. The sampling efficiency of quartz filters used in TOM is not well

34 characterized for small particles (Lim et al., 2014). However, smaller particles normally contribute little

35 to total particulate mass (Hinds 1999). Thus, each method for measuring BC in snow has both

36 advantages and disadvantages.

1   ~~In addition to BC, other LAI may contribute significantly to the radiative balance of the cryosphere.~~
2   ~~Recent research has identified mineral dust and microbiology as having a more important role than~~
3   ~~previously thought in the current decline in albedo of the Greenland Ice sheet and other parts of the~~
4   ~~Arctic (e.g. Dumont et al. 2014, Lutz et al., 2016). Similarly, Kaspari et al. (2014) reported such high~~
5   ~~dust concentrations in the snow of Himalayan Nepal that the contribution of dust in lowering the snow~~
6   ~~albedo sometimes exceeded that of BC. The importance of dust has also been illustrated from other~~
7   ~~regions, for example the Colorado Rockies, US, where dust causes a significantly earlier peak in runoff~~
8   ~~(Painter et al. 2007). In the Arctic, Doherty et al. (2010) suggest that 30 to 50 % of sunlight absorbed~~
9   ~~in the snowpack by impurities is due to non-BC constituents. Evidently, dust has an important role in~~
10   ~~the cryospheric radiative balance. Differentiating between the different impurities in the snow is not~~
11   ~~trivial, however, and requires more than one analytical technique (Doherty et al., 2016). Traditionally,~~
12   ~~dust in snow has been quantified by gravimetrically measuring filters (e.g. Aoki et al., 2006; Painter et~~
13   ~~al., 2012). Other methods have consisted of using a transmitted light microscopy (Thevenon et al.,~~
14   ~~2009), a microparticle counter to measure the insoluble dust (Ginot et al., 2014), or mass spectrometry~~
15   ~~(using iron as a proxy for dust) (Kaspari et al., 2014).~~

16   Here we present observations of LAI in snow from two glaciers in the Sunderdhunga valley in the
17   Indian Himalaya, which have not to our knowledge, been explored previously with respect to LAI in
18   snow. Using a measuring approach whereby the TOM is combined with a custom-built particle soot
19   absorption photometer (PSAP), we perform laboratory tests to provide a correct interpretation of the
20   results. Our Himalayan observations are further compared to samples from Arctic Finland for their LAI
21   content.

22   **2. Methodology**

23   **2.1 Snow sample collection and site characteristics**

24   *2.1.1 The Indian Himalaya*

25   Snow samples were collected in September of 2015, during the Indian post-monsoon season, from two
26   adjacent glaciers in the Sunderdhunga valley (Figure 1). Bhanolti and Durga Kot glaciers (N 30º 12', E
27   79º 51') are located in the state of Uttarakhand, India. Facing northeast the glaciers cover an elevation
28   range of about 4400-5500 m a.s.l. and are two small valley-type glaciers ~~contributing to~~ in the Ganges
29   hydrological basin. Since the glaciers are situated at a relatively low altitude, they are more likely to be
30   exposed to BC than other Himalayan glaciers ~~residing in~~ at a higher altitude, as BC has been shown to
31   decrease with altitude in other parts of the Himalaya (e.g. Kaspari et al., 2014; Ming et al., 2013; Yang
32   et al., 2015). The Sunderdhunga area does not have any major local pollution sources. Regionally,
33   however, the small towns of Bageshwar (~40 km S; population ~9000) and Almora (~70 km S;
34   population ~34000), may play a role. On a larger scale, the Sunderdhunga area is affected by the large-

1 scale emissions from the Indo-Gangetic Plain (IGP). Measurements of airborne BC and other aerosol

2 particles at Mukteshwar, a distance of ~90 km southwards at an altitude of 2200 m a.s.l., have shown a

3 clear seasonal pattern in atmospheric concentrations with emissions originating from the IGP

4 (Hyvärinen et al., 2011; Raatikainen et al., 2017). With a peak during the pre-monsoon season (March-

5 onset of monsoon), the BC loading has been reported to decrease by about 70 % at Mukteshwar during

6 the monsoon (Hyvärinen et al., 2011). Similarly, dust concentrations in the air have been shown to peak

7 during the pre-monsoon season at Mukteshwar (Hyvärinen et al., 2011). The pre-monsoon season, also

8 known as the "dust-season" in India, brings air masses from the Thar Desert transporting dust to the

9 Himalaya (Gautam et al., 2013). Dust from local sources has also been identified at Mukteshwar during

10 this season (Hyvärinen et al., 2011).

11 At Durga Kot glacier four snow pits with varying depths were dug at different elevations, while at

12 Bhanolti glacier one snow pit was dug (see table 1 for snow pits and sample details). Snow samples

13 were collected with a metal spatula in Nasco whirl-pak bags, and thereafter brought to the designated

14 base camp where the snow was melted and filtered. Since it was not possible to maintain the crucial

15 frozen chain for the snow samples during transport back to the laboratory this approach of melting in

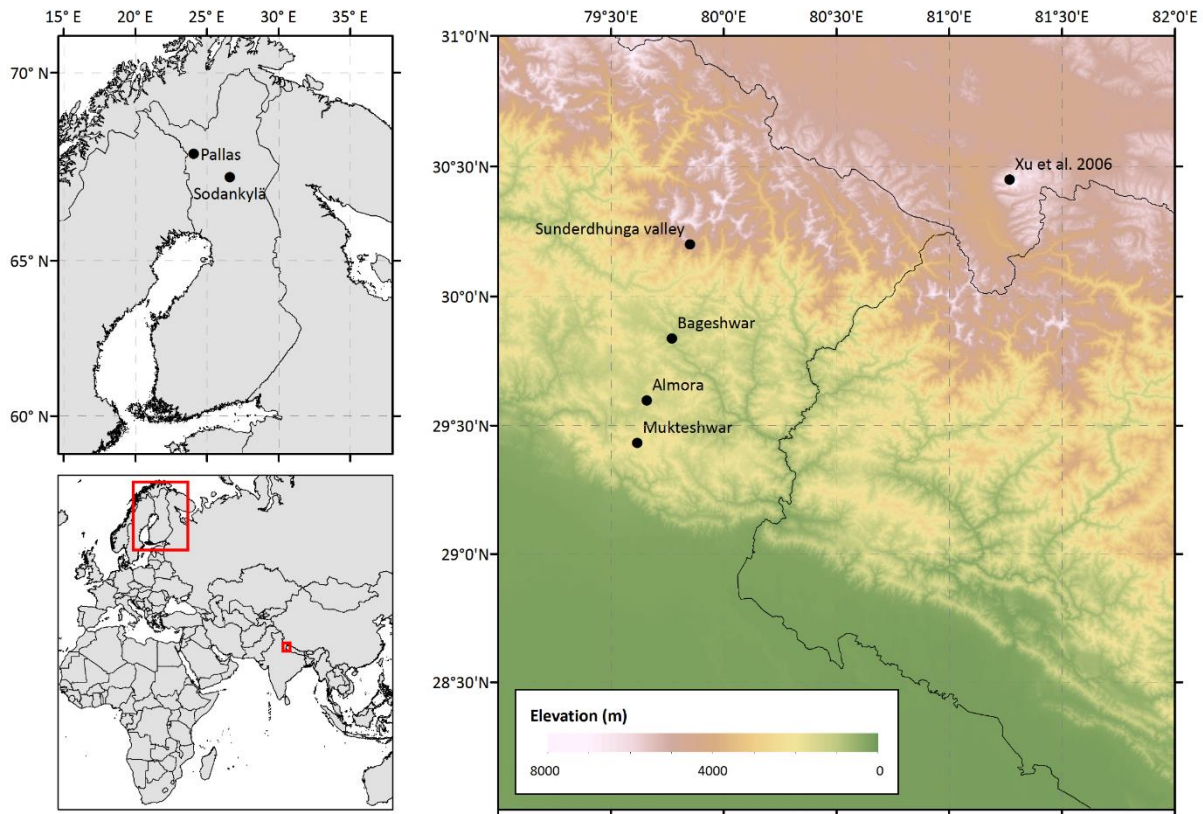
16 the field was used for the glacier snow samples. The snow was melted gently over a camping stove in

17 protected enclosed glassware to avoid contamination. The liquid samples were subsequently filtered

18 through quartz fiber filters (Munktell, 55 mm, grade T 293), in accordance to previous work (e.g.

19 Forsström et al., 2009; Svensson et al., 2013). The dried Filters were dried in ambient conditions in

20 petri dishes and thereafter then transported in petri dishes to the laboratory for analysis (described in

21 section 2.2).

2 Figure 1. ~~Google earth image~~ Map of ~~Indian~~ sampling locations and sites discussed in text.~~, as well as~~

3 ~~an overview map of measurement sites.~~

2.1.2 Arctic Finland

5 Snow samples collected in Finland originated from the seasonal snowpack of Sodankylä (N 67º 21' E

6 26º 37') and Pallas (N 67º 58' E 24º 06') c.f. Figure 1. The Pallas samples were gathered in March and

7 April of 2015 (n=10) from an open mire and in March of 2016 (n=2) from an area above the tree line

8 (in close proximity of the Pallas Global Atmosphere Watch Station). More details of the Pallas sampling

9 area are provided in Svensson et al. (2013) where EC in the snow was previously investigated. The

10 sampled snow was confined to the top layers of the snowpack. The Sodankylä samples (n=15) are from

11 the Finnish Meteorological Institute Arctic Research Center, where weekly surface snow samples (0-5

12 cm) have been collected since 2009 (the first part of time series is presented in Meinander et al., 2013;

13 where details of the area are provided). The samples used in this study originate from spring of 2013

14 and 2014. The snow samples from Pallas and Sodankylä were collected in Nasco whirl-pak bags and

15 stored in a frozen state until filtration. Samples were then melted in a microwave oven at each site's

16 respective laboratory, and followed the same filtering procedure described above, according to e.g.

17 Forsström et al. (2009) and Svensson et al. (2013).

18 **2.2 Light-absorbing impurities analysis**

1   To estimate the contribution to the reduction in transmission on the filter sample substrate due to

2   minerals, we compared the light transmission through the filter using the PSAP before and after heating

3   the sample as part of the TOM analysis. Since it is difficult to gravimetrically determine the dust content

4   on quartz filters, we decided to use this combined instrument approach to estimate the dust content. A

5   custom built PSAP (Krecl et al., 2007) was used for the optical measurements, and for the TOM a

6   Sunset Laboratory OCEC-analyzer was used to determine EC. A brief description of the OCEC-

7   analyzer and the PSAP is given below in sections 2.2.1 and 2.2.2, respectively.

8   The approach of measuring light transmission before and after heat treatment to estimate the different

9   light-absorbing components has been previously used for airborne sampled aerosol (e.g. Hansen et al.,

10   1993). In Hansen et al. (1993), filter samples were optically analyzed before and after being treated in

11   a 600ºC furnace, in which the carbonaceous material was vaporized from the filter. These measurements

12   enabled them to obtain an estimate of the dust content on the filter. Lavanchy et al. (1999) followed a

13   similar optical and thermal approach to determine the BC and dust content of ice core samples. For the

14   EC measurement they used a two-step combustion procedure by Cachier et al. (1989), and in between

15   the thermal treatment they used a modified version of an aethalometer to measure the attenuation of

16   light through the filter. Our experimental method is analogous to that of Lavanchy et al. (1999).

17   However, as a Sunset Lab. OCEC-analyzer and a custom built PSAP were readily available to us, this

18   instrument configuration was used in our study. Because results from this type of analysis may be very

19   instrument specific, a series of laboratory tests (described in section 2.3) were conducted to confirm

20   reliability of the method before ambient snow samples were measured. The analysis procedure for the

21   filters (outlined further in section 2.3) was the same for the laboratory samples and the ambient samples.

22   *2.2.1 Elemental carbon analysis*

23   From a 10 cm^2 filter sample area, separate punches of 1 cm^2 were taken and analyzed for organic carbon

24   (OC) and EC content using a Sunset laboratory OCEC-analyzer (Birch and Cary, 1996) with the

25   EUSAAR_2 analysis protocol (Cavalli et al., 2010). First, in a helium atmosphere, the filter punch is

26   heated at different temperature steps. In this phase OC is volatilized and detected by a flame ionization

27   detector (FID). During the second stage, oxygen is introduced, and EC is released from the filter through

28   combustion. To account for pyrolysis (darkening of the filter) occurring during the first step, a laser (at

29   a 632 nm wavelength) measures the transmittance (or reflectance as an option for newer instruments)

30   continuously of the filter punch, and when the original value of the transmittance (measured before

31   thermal sequence starts) is attained during the second step separation between OC and EC is done. The

32   ~~filters~~ EC values reported here (referred to as EC$_{TOM}$) are based on the transmittance correction for

33   pyrolysis since the PSAP operates also on the basis of transmittance through the substrate. An additional

34   EC value provided by the OCEC-analyzer from the analysis is an optical EC (EC$_{optical}$), which is based

35   on the monitored transmittance and absorption coefficients of the OCEC-analyzer. Carbonates may be

1   present in filter samples ~~For this study no special consideration was taken for carbon carbonate particles~~

2   ~~that can be present in the sample~~ (Chow & Watson, 2002), and ~~. U~~unless chemically removed before

3   analysis, these particles will contribute to the OC fraction of the total particulate carbon content in the

4   EUSAAR_2 protocol (~~e.g.~~ Cavalli et al., 2010). For our samples, carbonates are assumed to be present

5   in insignificant amounts since our visual inspection of the thermograms from analysis revealed no

6   carbonate peak. Recent tests by Zhang et al. (2017), on acidified and non-acidified filter samples with

7   snow from Tibet have showed less than 20 % discrepancy between treated and non-treated filter

8   substrates.

9   Uncertainties associated with the TOM method are mainly associated with ~~the inefficiency of the filters~~

10   ~~to capture the small impurities,~~ uneven filter loading, ~~and~~ loss of particles to filtering containers, and

11   the inefficiency of the filters to capture the impurities (undercatch) (Forsström et al., 2013; Lim et al.,

12   2014). For our filtering set-up the undercatch has been estimated to ca. 22 % (Forsström et al., 2013),

13   and is most likely significant for smaller sized particles, since undercatch tests have indicated an

14   inefficiency for smaller sized particles (Lim et al., 2014). During OCEC-analysis, ~~The~~ an artifact from

15   samples with a high fraction of pyrolysis OC (Lim et al., 2014), and the interference of an accurate split

16   point determination from filters containing a high dust load can also be considerable for the TOM

17   method (Wang et al., 2012).

18   *2.2.2 Absorption measurements*

19   The PSAP uses a single diode at 526 nm as light source. The light is split by two light pipes which

20   illuminate two areas of 3.1 mm in diameter. The filter substrate is placed over these areas and individual

21   detectors below the filter measure the transmitted light. During normal operations, when measuring BC

22   in air, these two signals are used as sample and reference spots. The reference spot is exposed to particle

23   free air and the sample spot is exposed to particles present in the ambient air. In this experiment both

24   signals are used to measure the change in transmission by comparing the signal before and after the

25   filter has been analyzed using TOM. The signal change is related to the transmission from a particle

26   free filter (filtered using Milli-Q (MQ) water and dried; the measurement procedure is further explain

27   in 2.3).

28   The corrections required for the PSAP when used for air sampling is well documented (e.g. Bond et al.,

29   1999; Virkkula et al., 2005), in particular this concerns enhanced absorption from the filter itself through

30   multiple scattering effects from the filter fibers, and particle loading effects (shadowing and reduction

31   in multiple scattering). However, these corrections are essentially uncharacterized for melted snow

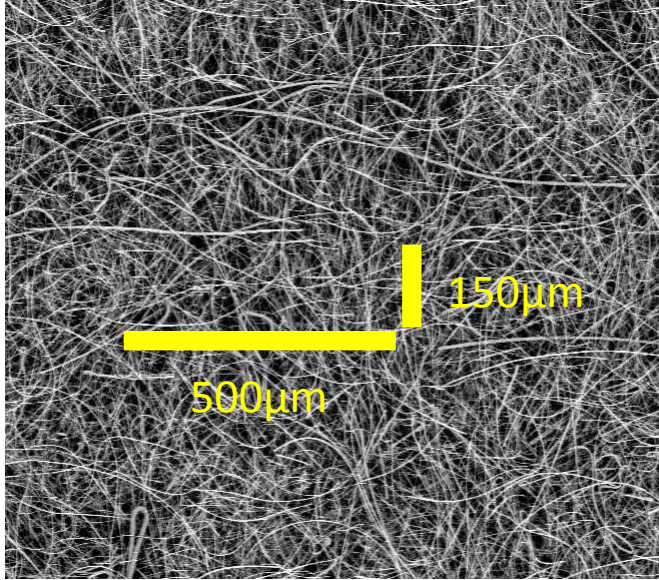
32   samples and the quartz fiber filters used. The fiber filters used are substantially thicker compared to

33   what is normally used for PSAP measurements (Pallflex cellulose membrane filter) or the ISSW

34   measurements (Nuclepore filter). Moreover, the filter substrate is very large in terms of surface area

35   compared to the particles sampled. The geometry is very complex and in relation to a particle the

1 substrate is more of a three dimensional web or sponge rather than a flat surface area on a filter. An
2 example of a blank filter sample obtained by a scanning electron microscope is presented in Figure 2.
3 The horizontal scale of 500 µm is for comparison, and the scale of 150 µm is to illustrate the relative
4 thickness of the substrate.

5

6 Figure 2. Electron microscope image of a blank quartz fiber filter used in this study.

7 The basis for the optical attenuation measurements is the exponential attenuation of light as it passes
8 through some medium, often described by the Bouguer-Lambert-Beer-law (Eq. 1).

9 $$I = I_0\, e^{-\tau}\,, \qquad \text{(Eq. 1)}$$

10 where I_0 in our case is the light intensity through a clean filter and, I is the light intensity through a
11 sample loaded filter. The exponent τ is the optical depth of LAI on the filter. For our study the multiple
12 scattering absorption enhancement factor of the filter will be treated as a constant, but not given a
13 numerical value. Due to the geometry of the filter, corrections for any enhanced absorption due to co-
14 existing scattering particles, and the loading effect, are not specifically considered. Hence, we will
15 assume a linear relation between the logarithmic change in transmittance (T_r) of a filter and the optical
16 depth (Eq. 2).

17 $$ln(T_r) = \tau_{TOT}, \text{ (Eq. 2)}$$

18 where $T_r = \frac{I_0}{I}$ and τ_{TOT} is the combined effect of all light absorbing impurities. Our interest was to
19 estimate the relative contributions of EC (τ_{EC},) and mineral dust (τ_D) particles to measured optical depth
20 according to equation 3.

21 $$\tau_{TOT} = \tau_{EC} + \tau_D, \text{ (Eq. 3)}$$

1   From TOM we get the EC mass surface density (µg cm^{-2}). Thus, we can write τ_{EC} as the product of the

2   EC mass surface density on the filter and an effective material specific mass absorption cross section

3   MAC$_{eff,EC}$ of BC that includes the multiple scattering enhancement of the filter, which is applicable for

4   our measurements and not necessarily as universal MAC values. Typically, MAC values are reported

5   in units of m^2 g^{-1}.

6   **2.3 Laboratory tests**

7   ~~Before initiating analysis of the field samples,~~ A series of laboratory tests using the OCEC-analyzer and

8   the PSAP combination were conducted before initiating analysis of the field samples. For this purpose,

9   the following filter sets were created:

10   1. A set of filter samples (n=36) with different amounts of BC. Two types of soot (BC) were used

11   and each was mixed (a small ~~minute~~ amount of soot not weighed) with MQ water and a small

12   amount of ethanol (to enable mixing of the BC particles in the liquid) in an ultra-sonic bath.

13   One soot type was collected by chimney cleaners in Helsinki, Finland, originating from oil-

14   based combustion, and has been used previously in soot on snow experiments (Svensson et al.,

15   2016). The second type was a product from NIST (National Institute of Standards and

16   Technology), which consists of diesel particle matter from industrial forklifts, NIST-2975.

17   From the BC stock solutions, different amounts of solution were taken out and diluted with

18   additional water for the same total volume of filtrate (ca. 0.5 L liquid). The newly created

19   mixture solution was thereafter filtered using the same filter procedure as the ambient snow

20   samples (described in 2.1.1).

21   2. The second set of filters (n=16) generated contained mineral dust only. Analogous to the soot

22   mixtures, two types of mineral were used. The first mineral was SiC, (manufactured by

23   Carborundum), mesh nr. 1200, corresponding to particles approximately < 1 µm in diameter

24   (Manufacturer). It is light grey with hints of blue in color and the amount of SiC added to the

25   MQ water was measured using a digital scale (resolution of 10 µg) before filtration. With the

26   known concentration of the mixture, we observed how much of the weighed mineral was

27   deposited on the filter during filtration to estimate losses. By comparing the whole filters before

28   and after filtration gravimetrically, these tests showed that 10 % or less of the mineral was lost

29   during filtering. The second type of mineral consisted of stone crush from a site in Stockholm,

30   Ulriksdal, likely to be mainly granite. A sieve mesh nr. 400 was used for this material, which

31   corresponds to mineral particles of approximately < 38 µm in diameter. Filters were prepared

32   according to the procedure given above for the other mineral (SiC).

33   3. The last set of laboratory solutions made contained various mixtures of SiC mineral and

34   chimney soot (n=30). These filters were treated in the same way as described above, with a soot

35   stock solution and a mineral weighed solution being mixed into one solution.

1   The procedure to analyze all three sets of filters samples was identical. After the filter substrates had

2   dried, one punch (1 cm^2) from the filter was put into the PSAP instrument to measure the transmission

3   across the filter in relation to a blank filter. This punch was taken for analyses of OC and EC content

4   using the OCEC-analyzer. After the TOM, and removal of the carbonaceous particles, this filter punch

5   was again analyzed in the PSAP. Hence, we acquired the transmission through the filter before heating

6   and after heating in comparison to a blank filter. We did tests where the same filter punch was used in

7   the PSAP instrument as well as the OCEC-analyzer, and compared this to ~~twin~~ identical samples that

8   were used only in each ~~separate~~ instruments. Both procedures provided the same result. Furthermore,

9   extensive tests were carried out using blank filters that had been subject to filtering of MQ water and

10   treated the same way as prepared samples and the ambient snow samples. No measurable EC could be

11   detected on these filters. It should be noted that part of the second set of the laboratory filters (stone

12   crush mineral) were analyzed with a different, but identical, PSAP and OCEC-analyzer at a different

13   laboratory (Stockholm University).

14   **3.   Results and discussion**

15   **3.1 Laboratory samples**

16   The change in optical depth as a function of analyzed EC using our two standard types of BC particles

17   (filter set nr. 1) is shown in Figure 3. Both materials behave optically similar and the slopes are within

18   15 % of each other, with chimney soot having a slope of 39.8 ± 1.5 m^2 g^{-1} and NIST soot 34.4 ± 1.8 m^2

19   g^{-1} (fits have been set to a fixed intercept at 0; $\pm$ refers to standard error of slope). Previous studies of

20   atmospheric airborne BC aerosol and its MAC with different filter-based absorption photometers are

21   numerous, while reported MAC values for BC in snow are very sparse. The MAC value of BC is

22   dependent o~~n~~f many factors, such as particle size, density, and refractive index, mixing state (i.e.

23   coating)~~, thus many influences on it~~. Reported airborne BC MAC values are lower than what we found

24   for the two soot standards (which were mixed in liquid solution to simulate similar conditions as for our

25   ambient snow samples). However, the MAC of air sample usually takes into account the multiple-

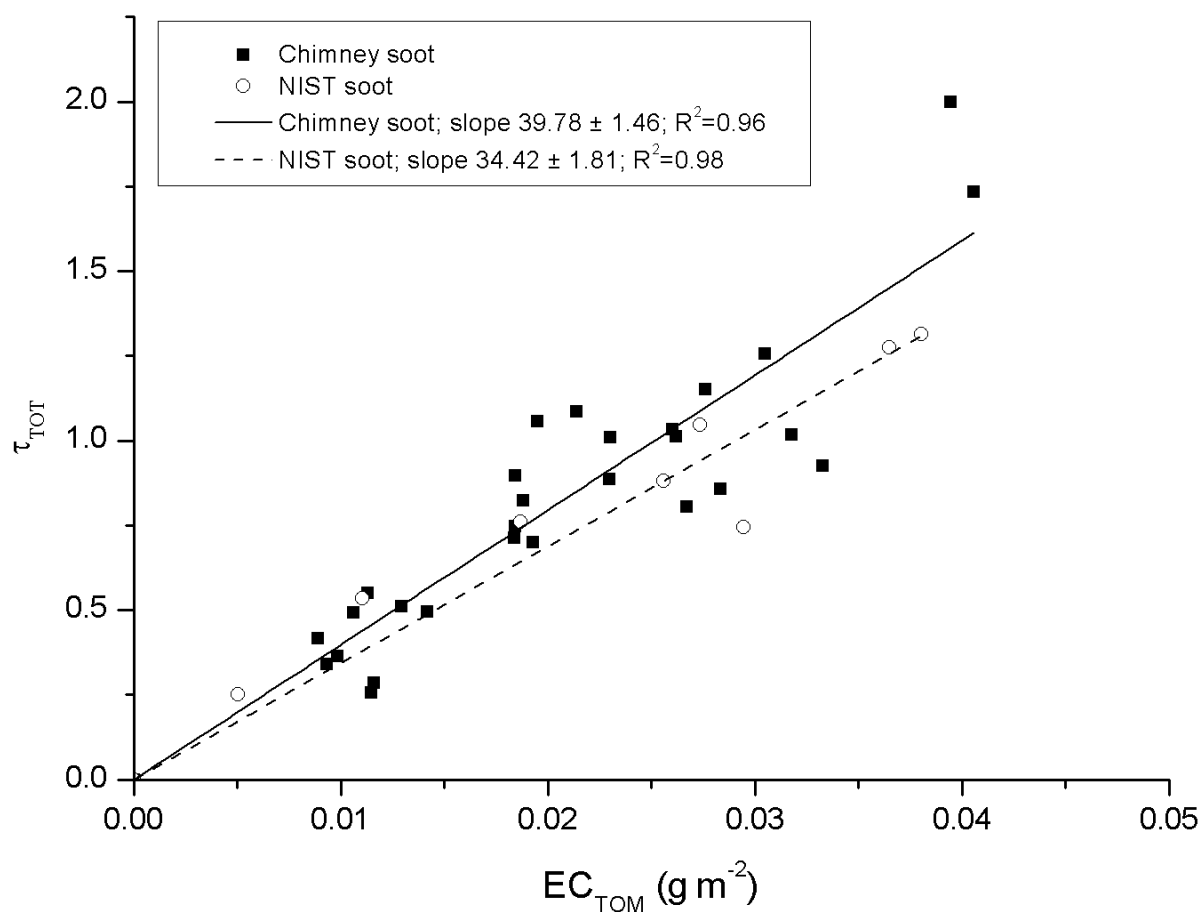
26   scattering correction factor (C_{ref}). For example for the commonly used aethalometer, its optical depth is

27   divided by a C_{ref} ~~somewhere~~ in the range of 2.8-4.3 (Collaud Coen et al., 2010). If a C_{ref} of 5.2 was

28   considered for our BC solution data ~~liquid originating BC data~~, similar MAC values would be found

29   (e.g. Bond et al. (2013) report~~s~~ freshly-generated BC with a MAC of 7.5 ± 1.2 m^2 g^{-1} at $\lambda = 550$ nm).

30   However, for our data set we have chosen not to take any C_{ref} into account as our samples are liquid

31   instead of air based, and currently no C_{ref} exists for liquid samples.

3   Figure 3. Comparison of ~~between~~ the optical depth (at λ=526 nm) by Chimney and NIST soot as

4   function of analyzed EC density by the OCEC-analyzer.

5   Figure 4 shows results ~~the~~ analogous ~~results as in~~ to Figure 3, but for the two mineral aerosol solutions

6   (filter set nr. 2). The slope of the optical depth of SiC versus measured SiC amount is more than a factor

7   of one hundred smaller (0.23 ± 0.008 m^2 g^{-1}) than the slopes for our BC standards. This is consistent

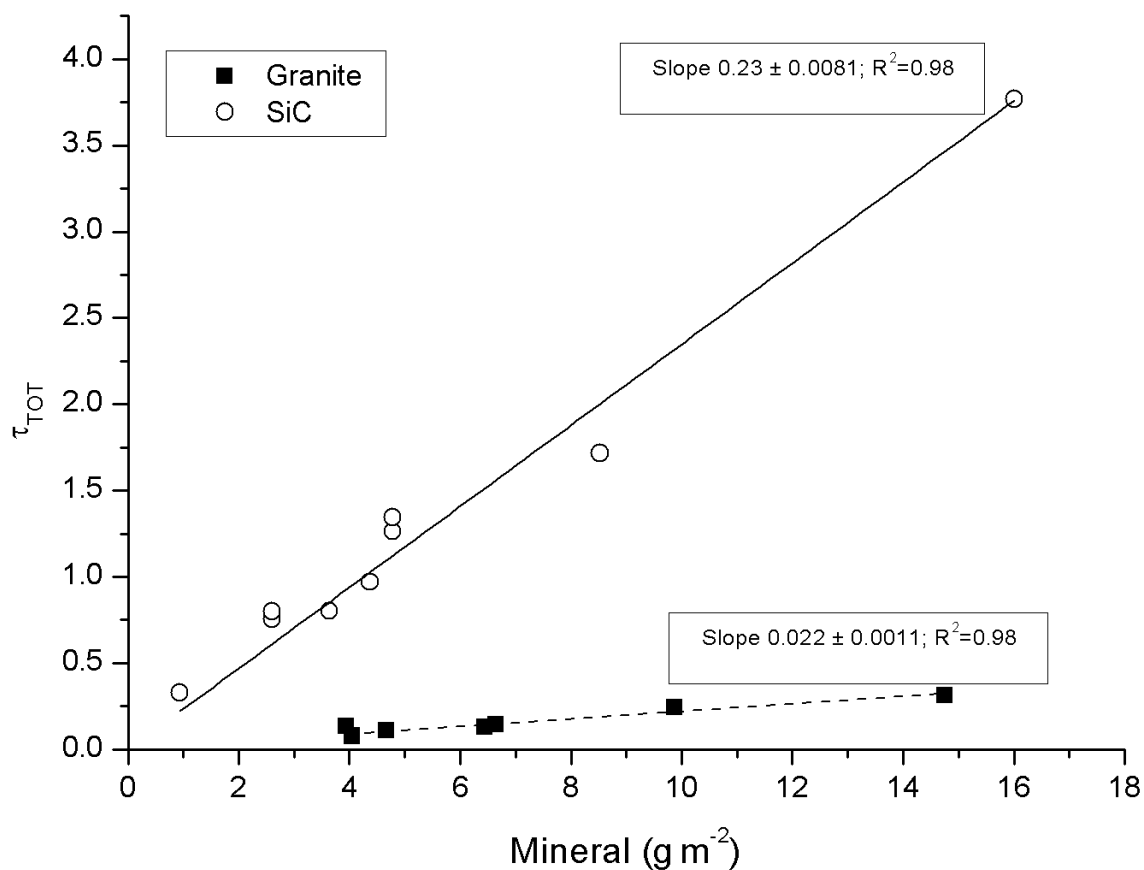
8   with previously reported results for airborne mineral dust (e.g. Hansen et al., 1993). The stone crush

9   material, an essentially white powder, yielded an even smaller slope of 0.02 ± 0.001 m^2 g^{-1}. Clearly, the

10 slopes, or the MAC, for the mineral particles are very composition specific. For a few (n=5) of the

11 mineral aerosol samples the optical depth was measured both before and after TOM. No EC was

12 detected on these samples and no significant difference in τ could be observed before and after heating

13 the sample, as one would expect since no BC was added to these filters.

2   Figure 4. The optical depth (at λ=526 nm) as a function of the amount of minerals present on the filter.

3   From the analysis of chimney and NIST soot (Fig. 3) and SiC and stone crush dust (Fig. 4) the

4   experiments were extended to comprise mixtures of soot and dust. Using the MAC of chimney soot

5   (see Fig. 3), we estimate the EC content of the third set of filters, containing a mixture of SiC and

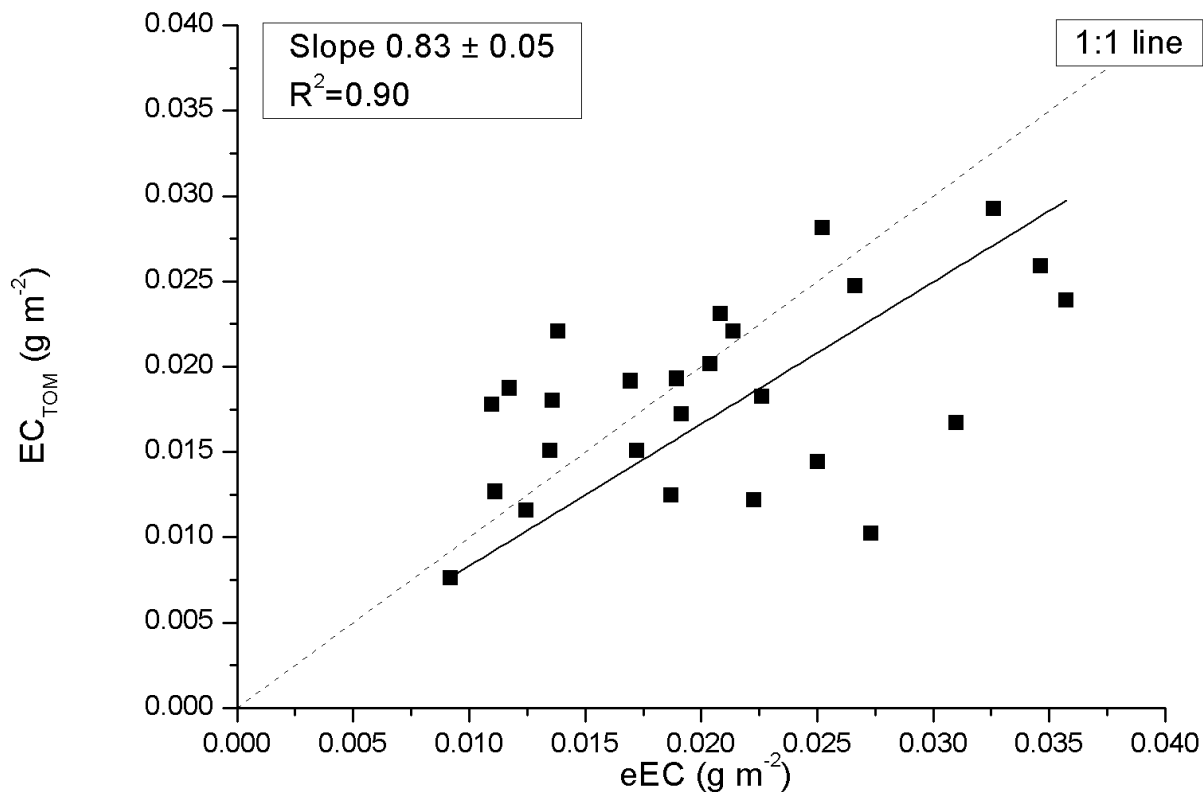
6   chimney soot. The estimated EC (eEC) is based on the difference between the optical thickness before

7   TOM analysis (τ_{TOT}) and the optical thickness after the analysis (τ_D). eEC is then compared to the

8   amount of EC obtained in TOM, for the same filters. This comparison is presented in Figure 5. The data

9   are is rather scattered, but the slope of the linear regression is within 17 % of the 1:1 line relatively close

10   (17 %) to 1:1. Hence it shows that EC can be reproduced reasonably well based on the PSAP

11   measurement even for a mixture of BC and minerals.

2   Figure 5. EC amount observed by the TOM (EC_{TOM}) for Chimney soot and SiC mixtures as a function

3   of estimated EC (eEC), using a PSAP optical depth signal before and after heating the filter and using

4   the $MAC_{eff,EC}$ of 39.8 m^2 g^{-1} from Figure 3.

5   In the context of this work it is further useful to compare our eEC content with the optical EC reported

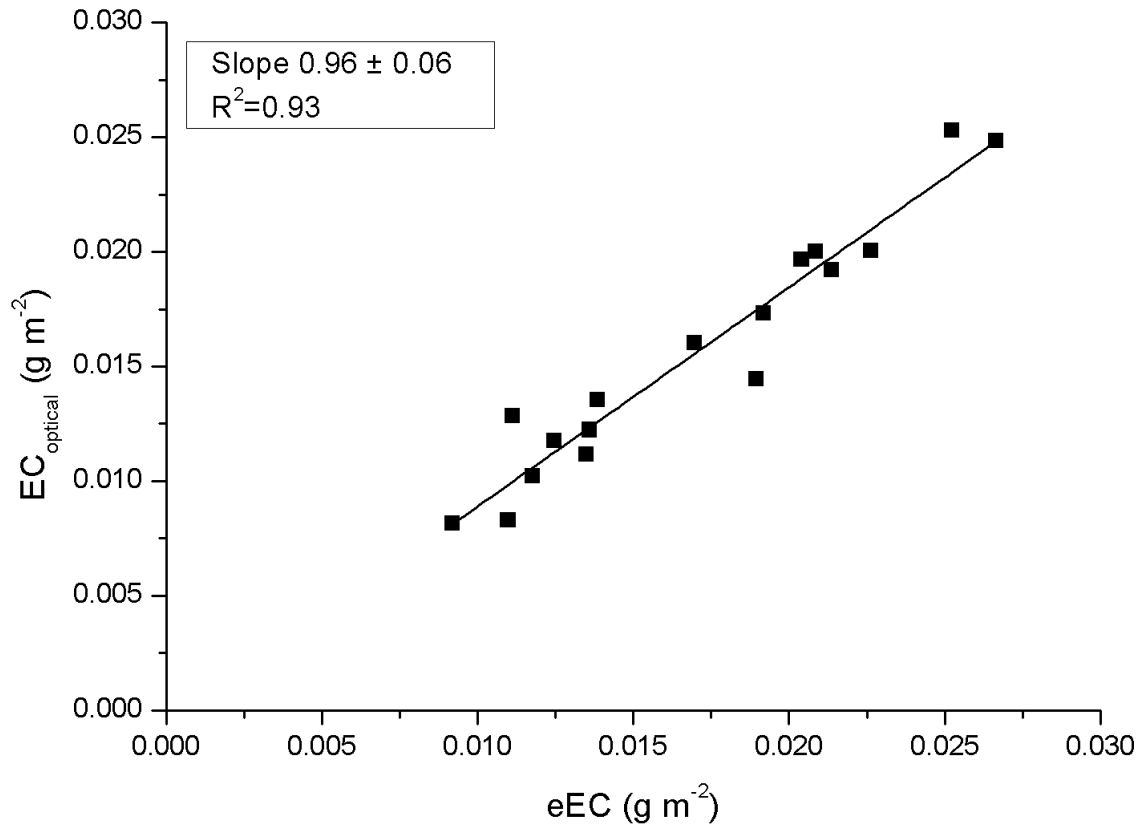
6   by the OCEC-analyzer. This comparison is presented in Figure 6, again for the third set of filters

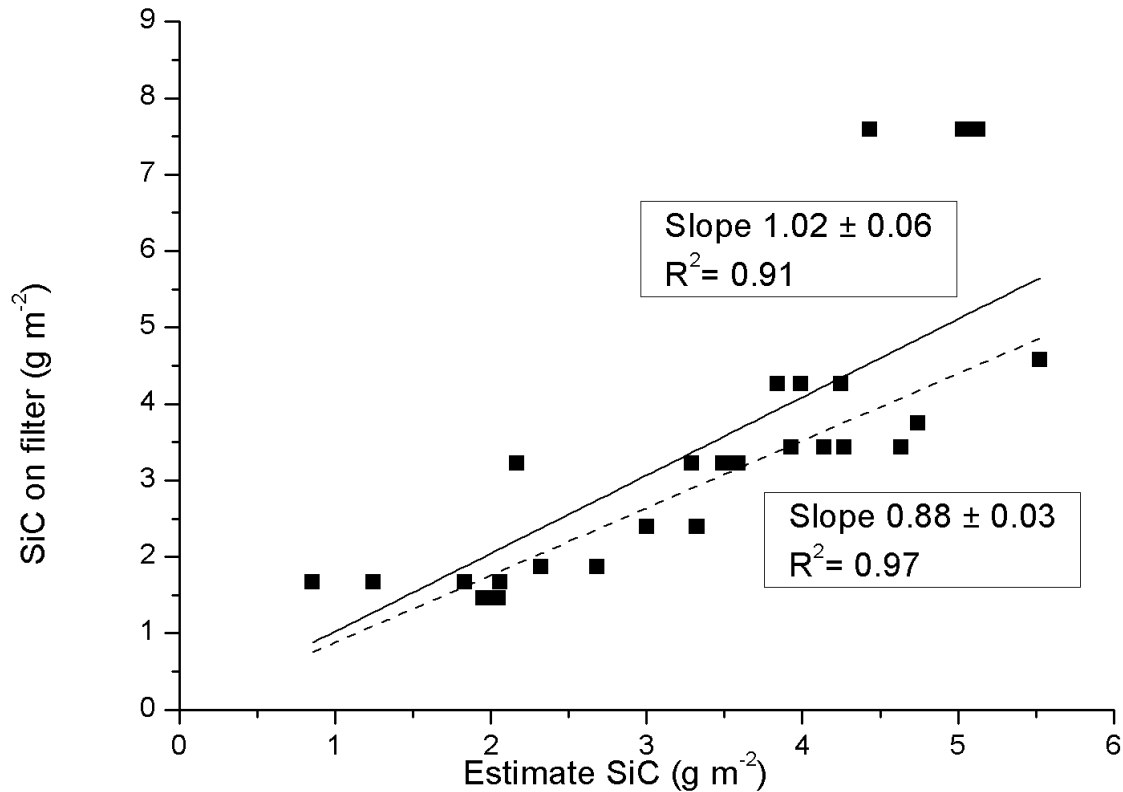
7   (Chimney soot+SiC). As observed, the EC amounts derived by two optically different methods show a

8   consistent relation ~~derived EC amounts show a very consistent relation~~ with nearly a slope of one. The

9   good agreement between the two optically derived EC values suggests that much of the scatter seen in

10   Figure 5 is due to the difference in the detection techniques (optical vs. TOM). The observed scatter in

11   Figure 5 could also be related to the fact that BC is mixed with SiC.

2   Figure 6. Comparison between the optically measurement of ~~reported~~ EC by the OCEC-analyzer and
3   the derived EC surface amount on the substrate ~~(~~using PSAP data and the relation in Figure 3~~)~~. The data
4   is for filters containing mixtures of Chimney soot and SiC.

5   In addition to chimney soot, the mineral SiC is the second absorbing component on the third set of
6   filters. In Figure 7 the optically estimated SiC content, based on the SiC slope in Figure 4 and τ_D is
7   compared to the known weighed amount of SiC before adding it to the liquid. Similarly, as in Figure 5,
8   there is some scatter in the data, but the overall pattern indicates a consistency with a reliable optical
9   measurement. ~~There are t~~wo slopes presented, one including all of the data points (slope 1.02), and the
10 second slope (0.88) excluding three data points with weighed SiC amounts exceeding 7.5 g m^{-2}.

2   Figure 7. Comparison between the weighed SiC amounts added to the water and the optically derived

3   SiC density on the substrate. The data is for Chimney soot and SiC mixtures, with two alternative slopes;

4   one containing all data points (1.02), and one excluding three data point in the top right of graph (0.88).

5   Based on the relations established for EC and SiC individually in figures 3 and 4, respectively, it is

6   possible to retrieve their separate concentrations from a mixture based on the change in filter

7   transmission before and after heating the filter. The consistent results from these laboratory tests gives

8   confidence in applying this method on our ambient samples from India and Finland.

9   **3.2 Ambient snow samples**

10   *3.2.1 EC in snow*

11   In all of the snow pits from Sunderdhunga a distinct layer with concentrated impurities was observed.

12   These impurity layers always had the highest EC concentrations (exceeding 300 µg L^{-1}) of each pit

13   (Table 1). For some of the samples from Sunderdhunga taken from the impurity concentrated layers,

14   the substrates were actually too loaded with material ~~that~~ and quantitative impurity values could not be

15   determined (by not having an initial transmission value). Excluding these heavy impurity layers, the

16   average and median EC concentration for the other snow samples were 141.3 and 101.9 µg L^{-1},

17   respectively. Surface samples taken above 4900 m a.s.l. had EC concentrations in the range of 13.2-

1   65.7 µg L^{-1}. Consisting of relatively fresh snow, fallen during the previous days (or weeks), these

2   surface samples contained LAI mostly ~~content is likely to~~ originat~~ing~~ from the post-monsoon season.

3   Previous studies of~~n~~ BC in snow and ice from the Himalaya have shown seasonal variation. At Mera

4   glacier in Nepal Ginot et al. (2014) showed that BC concentrations peak during the pre-monsoon in a

5   shallow ice core. From the same glacier, Kaspari et al. (2014) observed similar seasonal peaks in BC

6   concentration in snow and firn samples taken above the equilibrium line altitude, where the snow had

7   not undergone any significant summer melt. Noteworthy, dust did not show the same strong seasonality

8   as BC in their studies (Ginot et al., 2014; Kaspari et al., 2014).

9   Measurements of BC in snow taken closest to Sunderdhunga, reported in the literature, are from about

10 140 km east-north-east (78º heading), at a higher altitude between 5780-6080 m a.s.l. Gathered in the

11 surface snow of Namunani glacier Xu et al. (2006) reported low EC concentrations in the range of 0.3-

12 9.7 µg L^{-1}~~ng g^{-1}~~. The difference between Sunderdhunga and Namunani can probably be attributed to the

13 expected high spatial variability of BC in snow in the area. The difference in sampling altitude and

14 different measurement techniques to determine the EC likely plays a role as well (Xu et al. used a two-

15 step heating-gas chromatography, similar to method of Lavanchy et al.). The difference could also

16 possibly be explained by the geographical location, with Namunani located on the northern flank of the

17 Himalaya, and it is on the leeward side of the main sources of LAI to the south. Furthermore, it is not

18 explicitly stated in Xu et al. during which season snow samples were collected, which likewise would

19 affect EC concentrations.

20 For reference, the EC concentration in the surface ~~in relation to the comparison of the dust signal below~~

21 ~~in 3.2.2, the EC concentrations in the surface~~ snow from the Finnish Arctic were in the range of 6.2-

22 102 µg L^{-1}. Samples from Pallas had an average and median of 40.0 and 31.0 µg L^{-1}, respectively,

23 whereas the samples from Sodankylä had an average of 23.7 µg L^{-1} and median of 13.1 µg L^{-1}. The

24 higher concentration observed in Pallas might result from the fact that the ~~is likely because a~~ majority

25 of ~~the~~ samples was taken ~~originated from~~ later in the snow season compared to Sodankylä samples and

26 EC has likely concentrated in the surface snow later in the season (e.g. Svensson et al., 2013). On a

27 larger ~~broader~~ scale, the concentrations are in the same magnitude as previous measurements of EC in

28 snow from the European Arctic (Forsström et al., 2013; Meinander et al., 2013; Svensson et al., 2013).

29 Our snow samples EC content is further compared in Figure 8, where the estimated EC content based

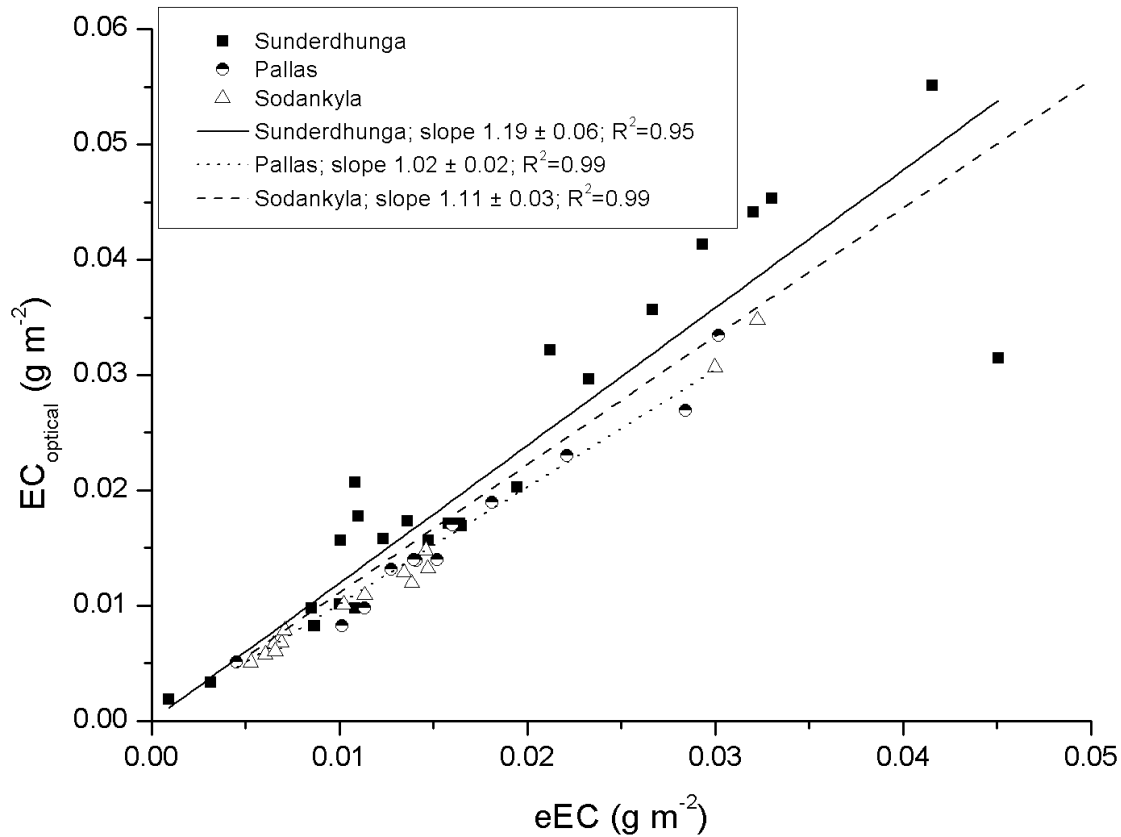
30 on the optical depth measurement is plotted against the optical EC output from the OCEC-analyzer. The

31 snow data presented in Figure 8 indicate the same relation between the two optical methods as presented

32 in Figure 6 for the standard soot. That is, slopes near 1:1 line, namely 1.19, 1.02, and 1.11 for

33 Sunderdhunga, Pallas, and Sodankylä samples, respectively. Hence, there is a strong consistency

1   between the two optical approaches in the interpretation of the change in τ before and after the substrate

2   has been analyzed with the EUSAAR-2 thermal protocol.

4   Figure 8. Comparison between the optical EC content given by the OCEC-analyzer and estimated EC

5   (eEC) content using a PSAP and a $MAC_{eff,EC}$ of 39.8 m^2 g^{-1}, for the Arctic and Himalayan samples.

6   Although the EC content determined by the optical method of the TOM and the eEC content based on

7   the PSAP and a MAC value (Figure 3) agree well, there is a significant difference in the site specific

8   derived MAC values. In Figure 9 the optical depth of EC ($τ_{EC}$) is plotted as a function of the analyzed

9   EC (with TOM) for all of the snow samples. The slopes for the three sampling sites are 21.0, 21.9 and

10 17.1 m^2 g^{-1} (Pallas, Sodankylä, and Sunderdhunga, respectively). These values are around half of what

11 the laboratory standard BC tests show (Fig. 3), indicating a smaller ~~less~~ absorption~~bing~~ efficiency for

12 the EC particles originating from the snow compared to the laboratory particles. This is unexpected, as

13 any non-EC absorbing material or even scattering particles mixed with EC would tend to increase the

14 MAC value compared to pure BC particles (e.g. Cappa et al., 2012; Bond et al., 2013). In our case,

15 ~~which~~ we would expect the MAC to be greater ~~to occur~~ for our snow originating EC particles. A

16 consequence of a lower MAC for the snow EC particles could be that the snow albedo reduction caused

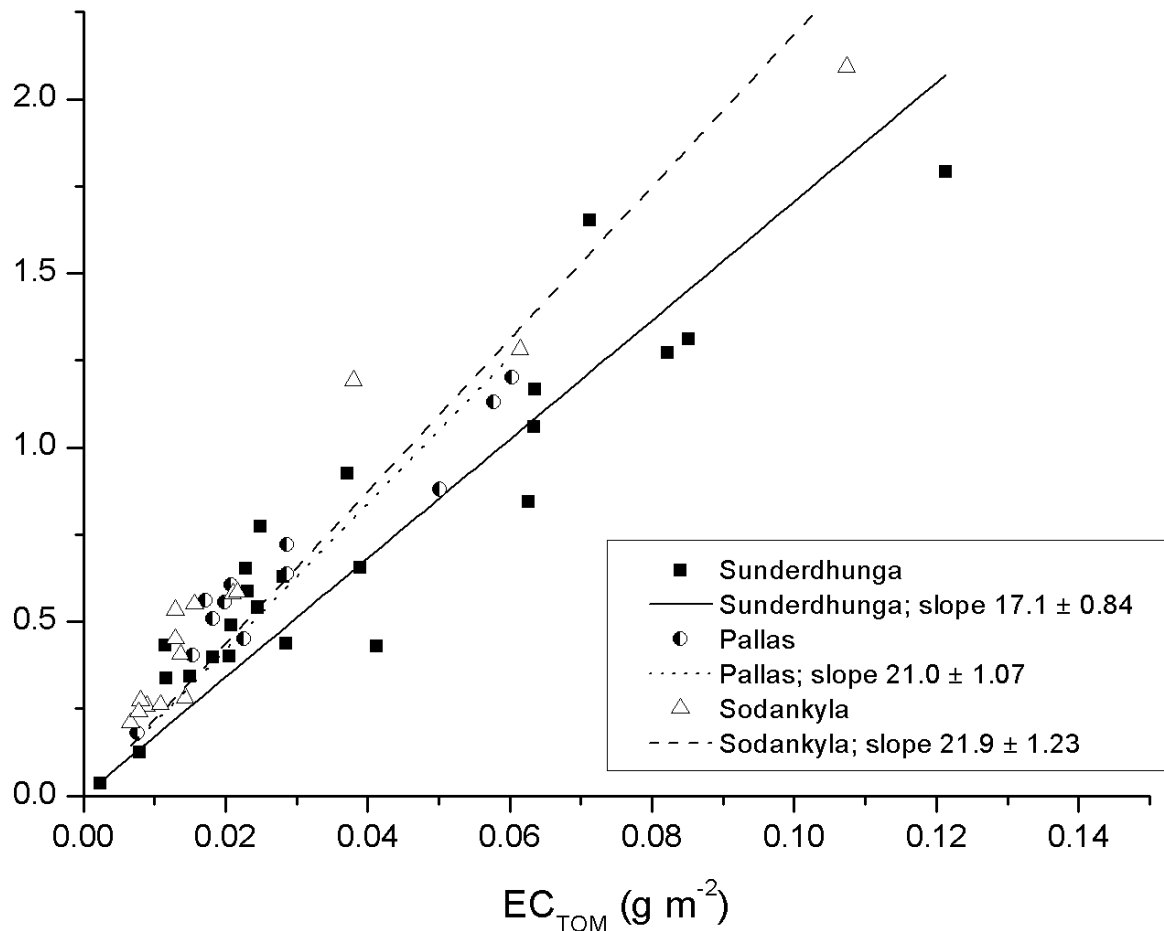
17 by the EC is inaccurate since the EC particles have less absorbing efficiency. Schwarz et al. (2013)

18 previously reported a lower MAC value for BC particles in the snow compared to airborne BC particles

1   due to a difference in the measured mean size. The BC particles from the snow were observed to be

2   larger compared to airborne BC particles, explaining the decrease in MAC for the snow ~~originating~~

3   particles. The authors further showed how the BC effect in snow albedo reduction is currently

4   overestimated due to the lower MAC for snow BC particles.

6   Figure 9. The optical depth τ_{EC} as function of the analyzed EC based on TOM, for the Arctic and

7   Himalayan samples.

8   In our case, if the laboratory generated BC consist of smaller particles compared to the snow samples

9   this could lead to a larger MAC value for the lab-standards. The size distribution of the BC particles in

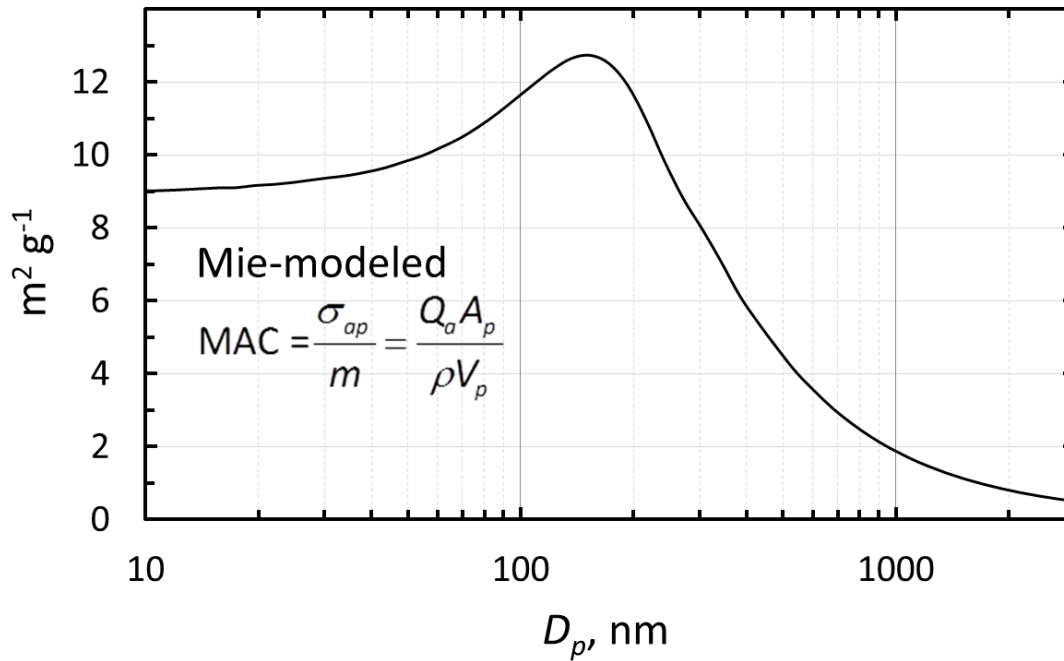
10   the filters are unknown to us, but as suggested by the modelled MAC curve, presented in Figure 10, this

11   size dependence can play a role. The modelled MAC for theoretical BC particles demonstrates a

12   decrease in MAC with particle size, particularly for particles larger than about 130 nm. The absorption

13   efficiencies were calculated for λ = 526 nm by using the Mie code of Barber and Hill (1990) and for

14   BC we use the same complex refractive index of 1.85 - 0.71i that was used by Lack and Cappa (2010)

15   and a particle density of 1.7 g cm^{-3}.

2   Figure 10. Modeled mass absorption coefficient (MAC) of single BC particles as a function of particle

3   diameter at $\lambda = 526$ nm.

4   Another hypothesis is related to the fact that the samples are liquids and that the matrix is strongly light

5   scattering and rather thick. It is likely that the liquid will embed the particles deeper into the filter than

6   what is typical for air samples (e.g. Chen et al., 2004). In air and on filter surfaces, BC mixed with a

7   scattering medium shows enhanced the absorption. On the samples presented in Table 1, about 90 to

8   95% of the carbon is water insoluble organic carbon, whereas the laboratory BC wasere essentially free

9   from OC. This difference could explain the lower MAC for the ambient samples if the net effect of the

10   added OC actually made the BC less efficient absorber in this particular matrix. Further tests are

11   required, however, to confirm this hypothesis.

12   *3.2.2 Dust fraction of LAI in snow*

13   Because the ambient mineral dust MAC value is unknown for our snow samples, it is not possible

14   applicable to use the SiC or stone crush MAC values to estimate the dust content on the filters. Instead,

15   we use the fraction of minerals (f_D) expressed in percent of the total optical thickness, $\left(\frac{\tau_D}{\tau_{TOT}} \; 100 \; \% \right)$ to

16   estimate the mineral aerosol contribution to the filter absorption (at $\lambda= 526$ nm). In our data set, there

17   is a systematic difference between the two Arctic sites and the Himalaya site (Fig. 11). For Pallas and

18   Sodankylä f_D is typically less than 20 %, whereas for Sunderdhunga f_D is typically much greater than

19   that, with modes peaking fractions reached at both ca. 35 and 65 %. For the Arctic, the values are

20   broadly in line with previous estimates on the amount of light absorption caused by LAI other LAI than

21   BC, i.e. 30-50 % (e.g. Doherty et al., 2010).

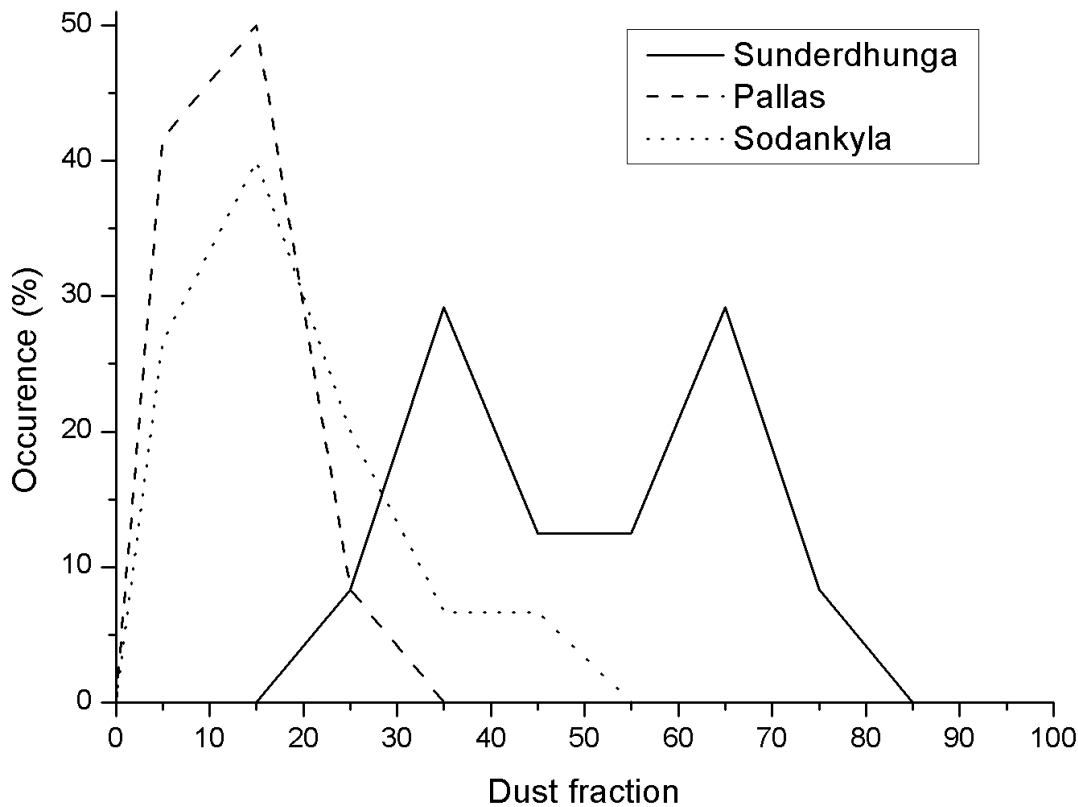
1   Studies from the Nepalese Himalaya concluded that dust may be responsible for about 40 % of the snow

2   albedo reduction (Kaspari et al., 2014). Similarly, Qu et al. (2014) observed that the contribution of dust

3   to albedo reduction can reach as ~~high~~ much as 56 % on a glacier ~~o~~in the Tibetan plateau. Our dust

4   estimate, as a fraction of the optical depth of LAI on the filter, shows similar results or an even greater

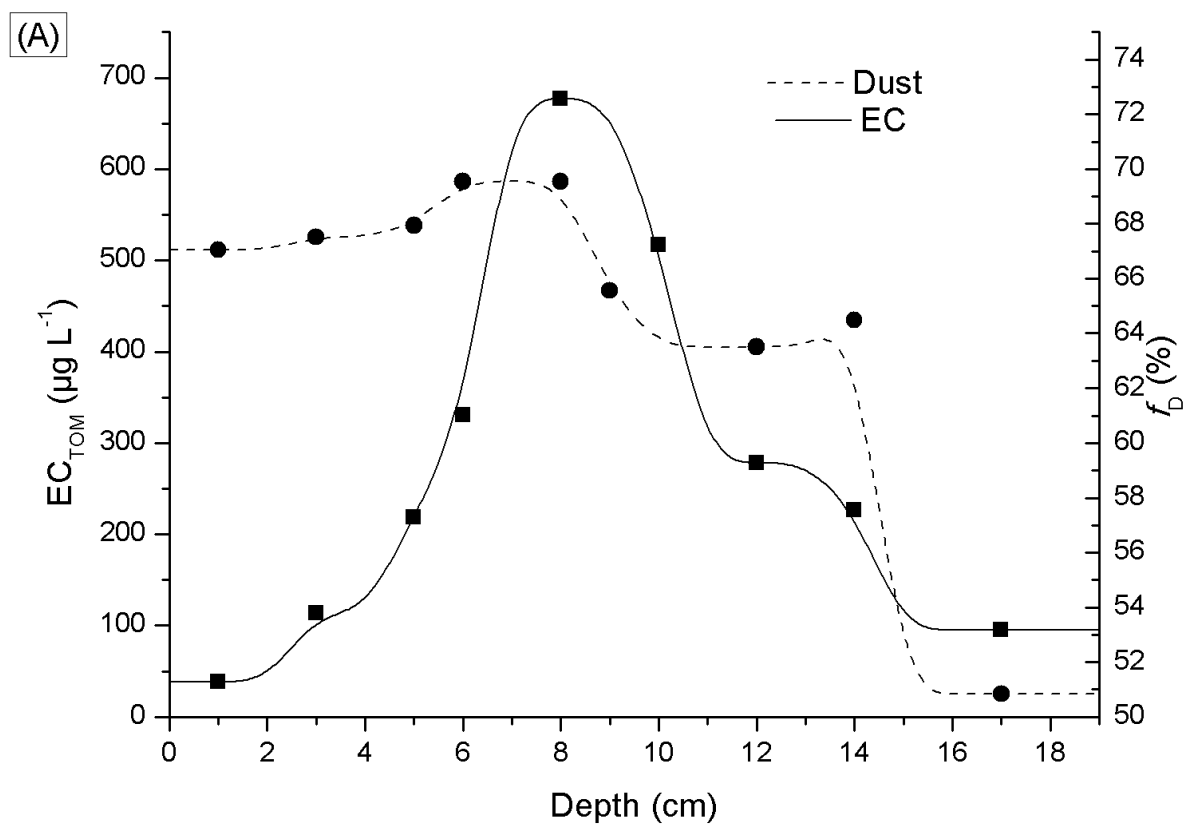
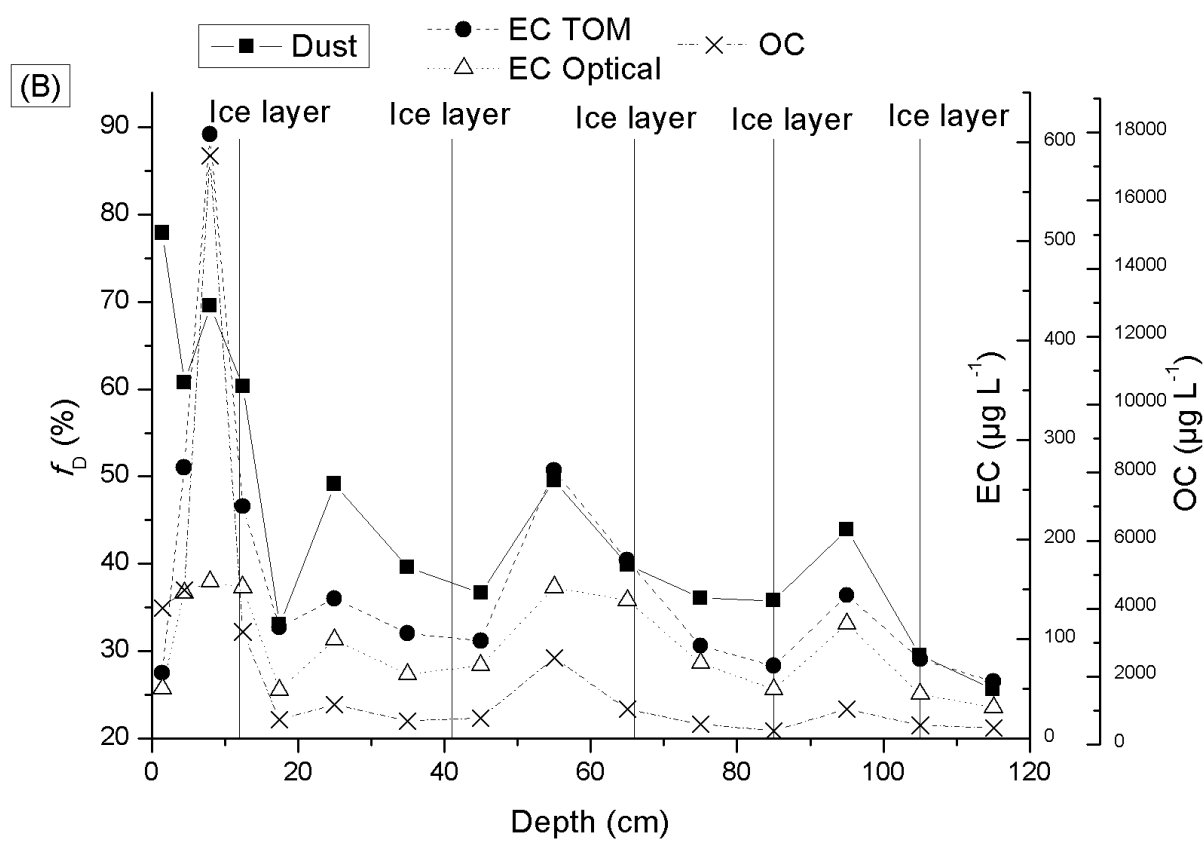
5   fraction of dust than these previous studies, highlighting the importance of dust (see also Fig. 12A)

6   causing ~~an~~ albedo reduction in this region of the Himalaya.

8   Figure 11. Frequency of the occurrence of dust optical thickness fractions at the three sampling sites.~~for~~

9   ~~different derived dust absorption fractions,~~ f_D.

10   *3.2.3   Vertical distribution of LAI in Sunderdhunga*

11   An average ~~composite~~ of the vertical profiles from pits C, D, and E are presented for EC and f_D in Figure

12   12A.

1 Figure 12. (A) Profile displaying average EC concentration and dust fraction from snow pits C and D

2 (Durga Kot glacier), and snow pit E (Bhanolti glacier); (B) Complete vertical profile E, taken at

3 Bhanolti glacier.

4 The variables plotted in Figure 12B display layers of enhanced amounts of both dust and EC, located

5 between ice layers, and additionally ~~evidently~~ high values at the top of the ~~core~~ pit above the first ice

6 layer. These layers are interpreted as indicators for seasonal variation at this location, with alternating

7 melt and refreezing periods marked by the ice layers. Since the ice layers and the enhancements in LAI

8 are interleaved it suggest that the impurities were deposited on the glacier mainly in-between the melt

9 and refreeze periods. In addition, the melting seems to take place in a relative shallow layer at the

10 surface and does not protrude deeply, which ~~would~~ may cause the annual layers to mix (Doherty et al.,

11 2013). The observed variation in EC and dust values could correspond to the findings of Ginot et al.

12 (2014) and Kaspari et al. (2014) who ~~that~~ showed annually peaking BC concentrations in the pre-

13 monsoon in Himalayan ice cores. However, for instance, between the ice layers at ca. 65 and 85 cm, no

14 clear peak is observed in EC or dust values (Fig. 12B), which could either indicate that no peak occurred

15 during that particular year, or an ice layer formed at ca. 65 cm in the middle of the year, similarly as

16 potentially at ca. 105 cm.

17 The snow pit covers ~~at best ca.~~ potentially five years of snow accumulation which is certainly a too

18 short time period to make any conclusions on a temporal trend of LAI variations at the site. However,

19 an evident increase in LAI is present, especially in the top 20 cm. Due to the time span of the snow pit

20 we cannot know for certain whether this increase presents a short term pollution event or indicates

21 increasing LAI at the site over a longer time period. We have two hypothesis for the observed increase

22 in EC concentrations and the fraction of dust occurring in the top layer of the snow pit. The higher

23 values may be a consequence of increased ambient EC and dust concentrations in the area, causing

24 increased dry and wet deposition fluxes of these impurities to the glacier, even when assuming constant

25 precipitation. Moreover, as it is f_D that increases, the deposition of dust would have had to increase

26 proportionally more than EC and OC. This could be a result from larger areas in the region being free

27 of snow or changes in the wind characteristics (e.g. stronger winds and/or change in direction). On the

28 other hand, local changes in the net snow mass balance due to a larger fraction of the snow being

29 sublimated in the time period covered by the top 20 cm in comparison to the deeper layers, may partly

30 explain the increased EC and dust absorption values at the top of the pit. Both these basic scenarios can

31 be in effect at the same time.

32 Interestingly, while the EC and OC concentrations and f_D are peaking at the top of the snow pit and

33 potentially decrease very slightly towards the bottom of the snow pit, the absorbing efficiency of EC

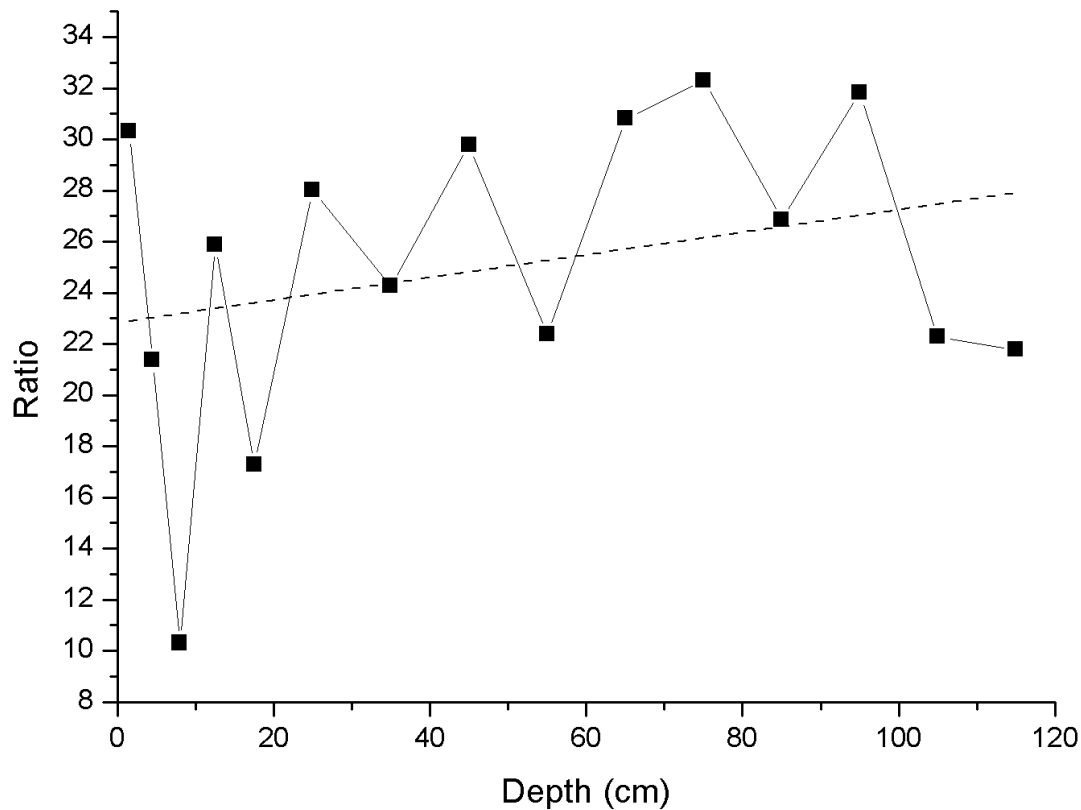
34 seems to be decreas~~ing~~ing towards the top of the snow pit. We illustrate this in Figure 13 by plotting the

35 ratio between the optical EC from the OCEC-analyzer and the analyzed EC based on TOM, and scale

1 this ratio with the MAC value of 39.8 derived in Figure 3. While the EC concentrations in the snow are

2 the highest at the top of the pit, it appears that at the same time this EC is a less potent light absorber

3 per unit mass (Fig. 13) than in deeper snow layers. This observation is rather hypothetical, and it needs

4 to be further explored.

6 Figure 13. The ratio between the optical EC content and analyzed EC content (TOM method) as

7 measured by the OCEC-analyzer using the EUSAAR-2 thermal protocol. The ratio is scaled by the

8 effective MAC value of 39.78 $m^2\ g^{-1}$ derived in Figure 3.

9 **4. Conclusions**

10 Here, first observations of LAI in snow originating from two glaciers in the Indian Himalaya are

11 presented with a method not used widely before to determine LAI in snow. Consisting of a custom built

12 PSAP and an OCEC-analyzer, the attenuation of light is studied on quartz filters, providing estimates

13 on the fraction of light-absorbance caused by non-EC constituents in LAI. Himalayan data display a

14 much greater light-absorbance by dust in the LAI compared to filter samples originating from the

15 seasonal snowpack of Arctic Finland. The role of dust in reducing the snow albedo in this part of

16 Himalayan glaciers needs to be further evaluated, as our results suggest that it might be the dominating

17 LAI in the snow. Our measurements further reveal that the optical properties of EC are different for

18 laboratory generated soot compared to EC deposited on ~~originating from~~ snow. Our finding of a ~~With~~

1   a MAC value off about half of the laboratory EC for the ambient EC particles, it can have potential

2   implications on for the snow albedo reduction caused by EC. Over the last approximately five year

3   period in the Himalaya, EC concentrations in the snow display signs of increase are elevated in the top

4   part of the snow pit compared to deeper layers, while at the same time its light absorbing potential

5   suggests a is decreaseing towards the highest EC-laden layers. Consequently, additional work on the

6   optical properties of EC in snow are needed to enable more accurate estimates of albedo reduction

7   caused by EC in snow, both spatially and temporally. This should be done by measuring the EC particles

8   light-absorption properties while in the snow since the ambient conditions can be different than

9   laboratory settings.

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21 **References**

22 AMAP: The Impact of Black Carbon on Arctic Climate. Arctic Monitoring and Assessment Programme
23 (AMAP), Oslo, Norway, 72 pp, 2011.
24 Aoki, T., Motoyoshi, H., Kodama, Y., Yasunari, T. J., Sugiura, K., and Kobayashi, H.: Atmospheric
25 aerosol deposition on snow surfaces and its effect on albedo. SOLA, 2, 13–16, doi: 10.2151/sola.2006-
26 004, 2006.
27 Barber, P. W. and Hill, S. C.: Light scattering by particles: Computational methods, World Scientific
28 Publishing, Singapore, 1990.
29 Birch, M. E., and Cary R. A.: Elemental carbon-based method for monitoring occupational exposures,
30 to particulate diesel exhaust, Aerosol. Sci. Technol., 25, 221– 241, 1996.
31 Bolch, T., Kulkarni, A., Kääb, A., Huggel, C., Paul, F., Cogley, J. G., Frey, H., Kargel, J. S., Fujita, K.,
32 Scheel, M., Bajracharya, S., and Stoffel, M.: The State and Fate of Himalayan Glaciers, Science, 336,
33 310–314, doi:10.1126/science.1215828, 2012.
34 Bond, T. C., Anderson, T. L., and Campbell, D.: Calibration and Intercomparison of Filter-Based
35 Measurements of Visible Light Absorption by Aerosols, Aerosol Sci. Technol. 30:582–600, 1999.
36 Bond, T. C., Doherty, S. J., Fahey, D. W., Forster, P. M., Berntsen, T., DeAngelo, B. J., Flanner, M. G,
37 Ghan, S., Kärcher, B., Koch, D., Kinne, S., Kondo, Y., Quinn, P. K., Sarofim, M. F., Schultz, M. G.,
38 Schulz, M., Venkataraman, C., Zhang, H., Zhang, S., Bellouin, N., Guttikunda, S. K., Hopke, P. K.,
39 Jacobson, M. Z., Kaiser, J. W., Klimont, Z., Lohmann, U., Schwarz, J. P., Shindell, D., Storelvmo, T.,
40 Warren, S. G., and Zender, C.S.: Bounding the role of black carbon in the climate system: A scientific
41 assessment, J. Geophys. Res. Atmos., 188, 5380–5552, doi: 10.1002/jgrd.50171, 2013.

Bond, T., Bhardwaj, E., Dong, R., Jogani, R., Jung, S., Roden, C., Streets, D. G., and Trautmann, N.: Historical emissions of black and organic carbon aerosol from energy-related combustion, 1850–2000, Global Biogeochem. Cy., 21, doi:10.1029/2006GB002840, 2007.

Cachier, H., Bremond, M.P., and Buat-Ménard, P.: Determination of atmospheric soot carbon with a simple thermal method, Tellus Ser.B, 41B, 379-390, 1989.

Cappa, C., Onasch, T., Massoli, P., Worsnop, D., Bates, T., Cross, E., Davidovits, P., Hakala, J., Hayden, K., Jobson, B., Kolesar, K., Lack, D., Lerner, B., Li, S., Mellon, D., Nuaaman, I., Olfert, J., Petäja, T., Quinn, P., Song, C., Subramanian, R., Williams, E., and Zaveri, R.: Radiative absorption enhancements due to the mixing state of atmospheric black carbon, Science, 337, 1078–1081, doi:10.1126/science.1223447, 2012.

Cavalli, F., Viana, M., Yttri, K.E., Genberg, J., and Putaud, J-P.: Toward a standardised thermal-optical protocol for measuring atmospheric organic and elemental carbon: the EUSAAR protocol, Atmos. Meas. Tech. 3, 79–89, doi:10.5194/amt-3-79-2010, 2010.

Chen, L.-W. A., Chow, J. C., Watson, J. G., Moosmüller, H., and Arnott, W. P.: Modeling reflectance and transmittance of quartz-fiber filter samples containing elemental carbon particles: Implications for thermal/optical analysis, J. Aerosol Sci., 35, 765–780, 2004.

Chow, J. C., & Watson, J. G.: PM2:5 carbonate concentrations at regionally representative Interagency monitoring of protected visual environment sites. J. Geophys. Res., 107(D21), 8344, doi:10.1029/2001JD000574, 2002.

Collaud Coen, M., Weingartner, E., Apituley, A., Ceburnis, D., Fierz-Schmidhauser, R., Flentje, H., Henzing, J. S., Jennings, S. G., Moerman, M., Petzold, A., Schmid, O. and Baltensperger, U.: Minimizing light absorption measurement artifacts of the Aethalometer: evaluation of five correction algorithms, Atmos. Meas. Tech., 3, 457–474, 10.5194/amt-3-457-2010, 2010.

Doherty, S. J., Warren, S. G., Grenfell, T. C., Clarke, A. D., and Brandt, R. E.: Light-absorbing impurities in Arctic snow, Atmos. Chem. Phys., 10, 11647–11680, doi: 10.5194/acp-10-11647-2010, 2010.

Doherty, S. J., Grenfell, T. C., Forsström, S., Hegg, D. L., Brandt, R. E., and Warren, S. G.: Observed vertical redistribution of black carbon and other insoluble light-absorbing particles in melting snow, J. Geophys. Res., 118, 1–17, doi:10.1002/jgrd.50235, 2013.

Doherty, S. J., D. A. Hegg, J. E. Johnson, P. K. Quinn, J. P. Schwarz, C. Dang, and Warren, S.G.: Causes of variability in light absorption by particles in snow at sites in Idaho and Utah, J. Geophys. Res. Atmos., 121, 4751-4768, doi:10.1002/2015JD024375, 2016.

Dumont, M., Brun, E., Picard, G., Michou, M., Libois, Q., Petit, J.-R., Geyer, M., Morin, S., and Josse, B.: Contribution of light-absorbing impurities in snow to Greenland's darkening since 2009, Nat. Geosci., 7, 509–512, doi:10.1038/ngeo2180, 2014.

Forsström, S., Ström, J., Pedersen, C. A., Isaksson, E., and Gerland, S.: Elemental carbon distribution in Svalbard snow, J. Geophys. Res. Atmos., 114, D19112, doi:10.1029/2008JD011480, 2009.

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.

Gautam R., Hsu, N. C., Lau, W. K.-M., and T. J. Yasunari, T. J.: Satellite observations of desert dust-induced Himalayan snow darkening, Geophys. Res. Lett., 40, 988–993, doi:10.1002/grl.50226, 2013.

Gertler, C.G., Puppala, S.P., Panday, A., Stumm, D., Shea, J.: Black carbon and the Himalayan cryosphere: a review. Atmos. Environ. 125, 404–417, doi.org/10.1016/j.atmosenv.2015.08.078, 2016.

Ginot, P., Dumont, M., Lim, S., Patris, N., Taupin, J.-D., Wagnon, P., Gilbert, A., Arnaud, Y., Marinoni, A., Bonasoni, P., and Laj, P.: A 10 year record of black carbon and dust from a Mera Peak ice core (Nepal): variability and potential impact on melting of Himalayan glaciers, The Cryosphere, 8, 1479–1496, doi:10.5194/tc-8-1479-2014, 2014.

Grenfell, T. C., Doherty, S. J., Clarke, A. D., and Warren, S. G.: Spectrophotometric determination of absorptive impurities in snow, Appl. Opt., 50(14), 2037–2048.

Hagler, G. S. W., Bergin, M. H., Smith, E. A., Dibb, J. E., Anderson, C., and Steig, E. J.: Particulate and water-soluble carbon measured in recent snow at Summit, Greenland, Geophys. Res. Lett., 34, L16505, doi:10.1029/2007GL030110, 2007.

1 Hansen, A.D. A., Kapustin, V. N., Kopeikin, V. M., Gillette, D. A., and Bodhaine, B. A.: Optical
2 absorption by aerosol black carbon and dust in a desert region of central Asia, Atmos. Environ., Part ,4,
3 27,4, 2527-2531, 1993

4 Hinds,W. C.: Aerosol Technology,Wiley-interscience, 1999.

5 Hyvärinen, A.-P., Raatikainen, T., Brus, D., Komppula, M., Panwar, T. S., Hooda, R. K., Sharma, V.
6 P., and Lihavainen, H.: Effect of the summer monsoon on aerosols at two measurement stations in
7 Northern India – Part 1: PM and BC concentrations, Atmos. Chem. Phys., 11, 8271-8282,
8 doi:10.5194/acp-11-8271-2011, 2011.

9 Immerzeel, W. W., van Beek, L. P. H., and Bierkens, M. F. P.: Climate Change Will Affect the Asian
10 Water Towers, Science, 328, 1382–1385, doi:10.1126/science.1183188, 2010.

11 Kaspari, S., Painter, T. H., Gysel, M., Skiles, S. M., and Schwikowski, M.: Seasonal and elevational
12 variations of black carbon and dust in snow and ice in the Solu-Khumbu, Nepal and estimated radiative
13 forcings, Atmos. Chem. Phys., 14, 8089–8103, doi:10.5194/acp-14-8089-2014, 2014.

14 Kääb, A., Berthier, E., Nuth, C., Gardelle, J., and Arnaud, Y.: Contrasting patterns of early 21st century
15 glacier mass change in the Himalaya, Nature, 488, 495–498, doi:10.1038/nature11324, 2012.

16 Krecl, P., Ström, J., and Johansson, C.: Carbon content of atmospheric aerosols in a residential area
17 during the wood combustion season in Sweden, Atmos. Environ., 41, 6974–6985, 2007.

18 Lack, D. A. and Cappa, C. D.: Impact of brown and clear carbon on light absorption enhancement,
19 single scatter albedo and absorption wavelength dependence of black carbon, Atmos. Chem. Phys., 10,
20 4207-4220, doi:10.5194/acp-10-4207-2010, 2010.

21 Lavanchy, V. M. H., Gäggler, H. W., Schotterer, U., Schwikowski, M., and Baltensperger, U.:
22 Historical record of carbonaceous particle concentrations from a European high-alpine glacier (Colle
23 Gnifetti, Switzerland), J. Geophys. Res., 104, 21227–21236, doi:10.1029/1999JD900408, 1999.

24 Lim, S., Faïn, X., Zanatta, M., Cozic, J., Jaffrezo, J.-L., Ginot, P., and Laj, P.: Refractory black carbon
25 mass concentrations in snow and ice: method evaluation and inter-comparison with elemental carbon
26 measurement, Atmos. Meas. Tech., 7, 3307-3324, doi:10.5194/amt-7-3307-2014.

27 Lutz, S., Anesio, A.M., Raiswell, R., Edwards, A., Newton, R.J., Gill, F., and Benning, L.G.: The
28 biogeography of red snow microbiomes and their role in melting arctic glaciers. Nat. Commun. 7:
29 11968, doi: 10.1038/ncomms11968, 2016.

30 Meinander, O., Kazadzis, S., Arola, A., Riihelä, A., Räisänen, P., Kivi, R., Kontu, A., Kouznetsov, R.,
31 Sofiev, M., Svensson, J., Suokanerva, H., Aaltonen, V., Manninen, T., Roujean, J.-L., and Hautecoeur,
32 O.: Spectral albedo of seasonal snow during intensive melt period at Sodankylä, beyond the Arctic
33 Circle, Atmos. Chem. Phys., 13, 3793-3810, doi:10.5194/acp-13-3793-2013, 2013.

34 McConnell, J. R., Edwards, R., Kok, G. L., Flanner, M. G., Zender, C. S., Saltzman, E. S., Banta, J. R.,
35 Pasteris, D. R., Carter, M. M., and Kahl, J. D. W.: 20th century industrial black carbon emissions altered
36 arctic climate forcing, Science, 317, 1381–1384, doi:10.1126/science.1144856, 2007.

37 Ming, J., Cachier, H., Xiao, C., Qin, D., Kang, S., Hou, S., and Xu, J.: Black carbon record based on a
38 shallow Himalayan ice core and its climatic implications, Atmos. Chem. Phys., 8, 1343– 1352,
39 doi:10.5194/acp-8-1343-2008, 2008.

40 Ming, J., Xiao, C., Du, Z., and Yang, X.: An Overview of Black Carbon Deposition in High Asia
41 Glaciers and its Impacts on Radiation Balance, Adv.Water Resour., 55, 80–87, 2013.

42 Ming, J., Wang, Y., Du, Z., Zhang, T., Guo, W., Xiao, C., Xu, X., Ding, M., Zhang, D., and Yang,
43 W.: Widespread albedo decreasing and induced melting of Himalayan snow and ice in the early 21st
44 century. PLoS One 10(6):e0126235. doi:10.1371/journal.pone.0126235, 2015.

45 Painter, T. H., Barrett, A. P., Landry, C. C., Neff, J. C., Cassidy, M. P., Lawrence, C. R., McBride, K.
46 E., and Farmer, G. L.: Impact of disturbed desert soils on duration of mountain snow cover, Geophys.
47 Res. Lett., 34, L12502, doi:10.1029/2007GL030284, 2007.

48 Painter, T. H., Skiles, S. M., Deems, J. S., Bryant, A. C., and Landry C.C.: Dust radiative forcing in
49 snow of the Upper Colorado River Basin: 1. A 6 year record of energy balance, radiation, and dust
50 concentrations. Water Resources Research, 48, doi: 10.1029/2012wr011985, 2012.

51 Qian,Y., Yasunari, T. J., Doherty, S. J., Flanner, M. G., Lau, W. K., Ming, J., Zhang, R.: Light-
52 absorbing particles in snow and ice: measurement and modeling of climatic and hydrological impact.
53 Adv. Atmos. Sci. 32(1),64–91, doi: 10.1007/s00376-014-0010-0, 2015.

1   Qu, B., Ming, J., Kang, S.-C., Zhang, G.-S., Li, Y.-W., Li, C.-D., Zhao, S.-Y., Ji, Z.-M., and Cao, J.-J.:
2   The decreasing albedo of the Zhadang glacier on western Nyainqentanglha and the role of light-
3   absorbing impurities, Atmos. Chem. Phys., 14, 11117-11128, doi:10.5194/acp-14-11117-2014, 2014.
4   Ruppel, M. M., Isaksson, E., Ström, J., Beaudon, E., Svensson, J., Pedersen, C. A., and Korhola, A.:
5   Increase in elemental carbon values between 1970 and 2004 observed in a 300-year ice core from
6   Holtedahlfonna (Svalbard), Atmos. Chem. Phys., 14, 11447-11469, doi:10.5194/acp-14-11447-2014,
7   2014.
8   Raatikainen, T., Brus, D., Hooda, R. K., Hyvärinen, A.-P., Asmi, E., Sharma, V. P., Arola, A., and
9   Lihavainen, H.: Size-selected black carbon mass distributions and mixing state in polluted and clean
10 environments of northern India, Atmos. Chem. Phys., 17, 371-383, doi:10.5194/acp-17-371-2017,
11 2017.
12 Schmitt, C. G., All, J. D., Schwarz, J. P., Arnott, W. P., Cole, R. J., Lapham, E., and Celestian, A.:
13 Measurements of light-absorbing particles on the glaciers in the Cordillera Blanca, Peru, The
14 Cryosphere, 9, 331-340, doi:10.5194/tc-9-331-2015, 2015.
15 Schwarz, J. P., Doherty, S. J., Li, F., Ruggiero, S. T., Tanner, C. E., Perring, A. E., Gao, R. S., and
16 Fahey, D. W.: Assessing Single Particle Soot Photometer and Integrating Sphere/Integrating Sandwich
17 Spectrophotometer measurement techniques for quantifying black carbon concentration in snow,
18 Atmos. Meas. Tech., 5, 2581–2592, doi:10.5194/amt-5-2581-2012, 2012.
19 Schwarz, J. P., Gao, R. S., Perring, A. E., Spackman, J. R., and Fahey, D.W.: Black carbon aerosol size
20 in snow, Nat. Sci. Reports, 3, 1356, doi:10.1038/srep01356, 2013.
21 Shindell, D., Kuylenstierna, J. C. I., Vignati, E., van Dingenen, R., Amann, M., Klimont, Z., Anenberg,
22 S. C., Muller, N., JanssensMaenhout, G., Raes, F., Schwartz, J., Faluvegi, G., Pozzoli, L., Kupiainen,
23 K., Höglund-Isaksson, L., Emberson, L., Streets, D., Ramanathan, V., Hicks, K., Oanh, N. T. K., Milly,
24 G., Williams, M., Demkine, V., and Fowler, D.: Simultaneously Mitigating Near-Term Climate Change
25 and Improving Human Health and Food Security, Science, 335, 183–189, doi:
26 10.1126/science.1210026, 2012.
27 Svensson, J. Ström, J., Hansson, M., Lihavainen, H., and Kerminen, V.-M.: Observed metre scale
28 horizontal variability of elemental carbon in surface snow, Environ. Res. Lett., 8, 034012,
29 doi:10.1088/1748-9326/8/3/034012, 2013.
30 Svensson J., Virkkula A., Meinander O., Kivekäs N., Hannula H.-R., Järvinen O., Peltoniemi J.I.,
31 Gritsevich M., Heikkilä A., Kontu A., Neitola K., Brus D., Dagsson-Waldhauserova P., Anttila K.,
32 Vehkamäki M., Hienola A., de Leeuw G. & Lihavainen H. 2016: Soot-doped natural snow and its
33 albedo — results from field experiments. Boreal Env. Res. 21: 481–503.
34 Thevenon, F., Anselmetti, F. S., Bernasconi, S. M., and Schwikowski, M.: Mineral dust and elemental
35 black carbon records from an Alpine ice core (Colle Gnifetti glacier) over the last millennium, J.
36 Geophys. Res., 114, 102, doi: 10.1029/2008JD011490, 2009.
37 Virkkula, A., Ahlquist, N. C., Covert, D. S., Arnott,W. P., Sheridan, P. J., Quinn, P. K., and Coffman,
38 D. J. (2005). Modification, Calibration and a Field Test of an Instrument for Measuring Light
39 Absorption by Particles, Aerosol Sci. Technol. 39:68–83.
40 Warren, S. G., and Wiscombe, W. J.: A model for the spectral albedo of snow. II: Snow containing
41 atmospheric aerosols, J. Atmos. Sci., 37, 2734–2745, 1980.
42 Wang, M., Xu, B., Zhao, H., Cao, J., Joswiak, D., Wu, G., and Lin, S.: The Influence of Dust on
43 Quantitative Measurements of Black Carbon in Ice and Snow when Using a Thermal Optical Method,
44 Aerosol Sci. Technol., 46, 60–69, doi:10.1080/02786826.2011.605815, 2012.
45 Xu, B., Yao, T., Liu, X., and Wang, N.: Elemental and organic carbon measurements with a two-step
46 heating-gas chromatography system in snow samples from the Tibetan Plateau, Ann. Glaciol., 43, 257–
47 262, 2006.
48 Xu, B., Cao, J., Hansen, J., Yao, T., Joswiak, D.R., Wang, N., Wu, G., Wang, M., Zhao, H., Yang, W.,
49 Liu, X., and He, J.: Black soot and the survival of Tibetan glaciers. Proc. Nat. Acad. Sci. USA, 106,
50 22114–22118, doi:10.1073/pnas.0910444106, 2009.
51 Yang, S., Xu, B., Cao, J., Zender, C. S., and Wang, M.: Climate effect of black carbon aerosol in a
52 Tibetan Plateau glacier, Atmos. Environ., 111, 71–78, doi.org/10.1016/j.atmosenv.2015.03.016 1352-
53 2310, 2015.
54 Zhang, Y., Kang, S., Li, C., Gao, T., Cong, Z., Sprenger, M., Liu, Y., Li, X,., Guo, J., Sillanpää, M.,
55 Wang, K., Chen, J., Li, Y., Sun, S.: Characteristics of black carbon in snow from Laohugou No. 12

1 glacier on the northern Tibetan Plateau, Sci. Tot. Environ., 607-608, 1237-1249,
2 doi.org/10.1016/j.scitotenv.2017.07.100, 2017.

1   Table 1. Snow pit filter samples from Sunderdhunga 2015. Durga kot glacier snow pits are A-D and Bhanolti glacier snow pit E.

Snow pit ID and elevation (m a.s.l.)	Sample interval (cm)	τTOT	τD	τEC	EC TOM (g m^{-2})	EC optical (g m^{-2})	eEC (g m^{-2})	Total C (g m^{-2})	EC (μg L^{-1})	F$_D$
A, 4869	0-2	3.94	2.63	1.31	0.09	0.05	0.03	1.75	362.18	66.68
	2-5	-	-	-	0.25	0.00	-	0.25	1010.62	-
	5-10	-	-	-	0.30	0.01	-	11.11	1030.84	-
B, 4921	0-2	0.69	0.26	0.43	0.01	0.01	0.01	0.15	40.33	37.64
	2-6	-	4.79	-	0.13	0.04	-	4.24	398.60	-
C, 4921	0-3	0.29	0.16	0.13	0.01	0.00	0.00	0.21	35.71	56.62
	3-6	1.76	1.32	0.44	0.03	0.02	0.01	1.24	55.15	75.12
	6-9	-	5.19	-	0.15	0.00	-	4.88	1095.79	-
	9-13	2.20	1.35	0.84	0.06	0.03	0.02	0.76	381.63	61.57
D, 4950	0-5	0.11	0.07	0.04	0.00	0.00	0.00	0.06	13.20	66.65
	5-10	-	-	-	0.07	0.01	-	6.70	327.14	-
	10-20	1.37	0.94	0.43	0.04	0.02	0.01	0.64	220.21	68.63
	20-30	1.15	0.66	0.49	0.02	0.02	0.01	0.34	78.58	57.36
E, 5008	0-3	1.81	1.41	0.40	0.02	0.02	0.01	1.27	65.73	77.87
	3-6	3.25	1.97	1.27	0.08	0.04	0.03	1.45	272.57	60.77
	6-10	5.88	4.09	1.79	0.12	0.03	0.05	3.58	607.83	69.55
	10-15	2.94	1.77	1.17	0.06	0.04	0.03	0.97	233.21	60.32
	15-20	0.98	0.32	0.66	0.04	0.02	0.02	0.30	111.42	33.05
	20-30	1.06	0.52	0.54	0.02	0.02	0.01	0.23	140.71	49.15
	30-40	1.04	0.41	0.63	0.03	0.02	0.02	0.21	105.55	39.61
	40-50	1.03	0.38	0.65	0.02	0.02	0.02	0.21	98.15	36.71
	50-60	2.10	1.04	1.06	0.06	0.04	0.03	0.66	269.67	49.53
	60-70	2.75	1.10	1.65	0.07	0.06	0.04	0.49	179.30	39.90
	70-80	1.21	0.44	0.77	0.02	0.02	0.02	0.19	93.00	36.12
	80-90	0.91	0.33	0.59	0.02	0.02	0.01	0.15	72.80	35.81

90-100	1.65	0.73	0.92	0.04	0.03	0.02	0.31	143.80	43.95
100-110	0.57	0.17	0.40	0.02	0.01	0.01	0.15	79.31	29.54
110-120	0.46	0.12	0.34	0.02	0.01	0.01	0.15	56.91	25.68