Interactive comment on “Seasonal and elevational variations of black carbon and dust in snow and ice in the Solu-Khumbu, Nepal and estimated radiative forcings” by S. Kaspari et al.

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Below we have copied the comments from referee #2. Our responses are embedded below, and are marked by a hyphen (-).

The authors describe measurements of BC and iron in snow samples collected in the high altitude area of the Khumbu valley in Nepal (two crevasse profiles and four snow pits from the Mera glacier, nine fresh snow samples from the Pyramid NCO-P station). The samples were analyzed using a combined nebulizer-SP2 system to determine BC concentrations, Fe was determined using ICP-MS. Furthermore, dust was determined in some samples using a gravimetric method. The authors describe in detail the uncertainties and limitations in the detection of BC in the samples including the uncertainty introduced due to the melting and storage of the melted samples and retain that the reported BC concentrations are possibly largely underestimated. In contrast, the two (!) reported dust concentrations are a factor of \( \times 8000 \) and \( \times 35000 \) higher than the measured BC concentrations in the same samples. If these concentrations are correct the impact of dust on the snow albedo and all further implications on the melting of snow and ice in this region is probably overwhelming (as even indicated by some limited calculations in the manuscript, Fig. 4). However, a detailed description of the detection method including an estimate of the uncertainty of the dust concentrations is missing. If the dust concentrations are correct, which is difficult to judge with the limited information provided by the authors, it would rectify our current understanding regarding the role of BC, dust, and other absorbing impurities in the snow for this sensible region. Since it has been demonstrated that the cryosphere and its behavior in this region may have important implications for the regional climate, glacier mass balance, and water resources in this region, such information would definitely warrant publication. Instead, the authors present only a number of general conclusions and remarks regarding the impact of absorbing impurities in the snow on the cryosphere without adding further many new findings.

-We have expanded on the discussion of the dust data, including providing further evidence in support of the gravimetric mass being dominated by dust, sources of uncertainty in the dust data, and the relative importance of dust vs. black carbon. All of the samples were analyzed for Fe via ICP-MS, so the claim that only two dust values are reported is inaccurate. Only samples with high impurity loads were measured using the gravimetric method because the lower concentration samples didn’t have enough mass to make the measurement. There were five samples that had high enough impurity concentrations to be measured gravimetrically. The assumption that the gravimetric dust measurement reflects dust is supported by:
1. The dry mass impurity is well correlated with the Fe measurements (Figure 1), suggesting that dust dominates the dry mass.

2. The ratio between the total solid mass and Fe mass is 1.25%, or 1.8% when considering Fe$_2$O$_3$, which is in the range expected for the upper continental crust (4.4%), granites (2%) and sedimentary rocks (6.45%) (Wedepohl et al., Geochimica et Cosmochimica Acta 59, 1217-1232, 1995). These calculations are based on the slope in Figure 1 8xE-05 g/l/ng/g, which is equivalent to 8xE-05 g/l/E-06 g/L (assuming density of 1 g/ml for the liquid sample used for Fe analysis). Thus the slope is 80, i.e. 80 g of dust per 1 g of Fe which is 1.25% Fe. Scaled to hematite (see details under point 3 below) results in 1.8% (see details under point 3 below on hematite conversion).

3. Takeuchi et al. (2002) reported that organic material accounted for between 3.0-6.8% of cryoconite mass on Himalayan glaciers, supporting the assumption that dust dominates the impurity mass (solids). We’ve expanded the discussion of the relative absorption of BC versus dust by using the BC and Fe data. Fe is a good dust proxy because iron oxides dominate light absorption by mineral dust. The fraction of the Fe that is light absorbing is estimated to be 50-65% of the measured Fe (Fairbridge et al., 1972) and in the form of iron oxides. The mass absorption cross section (MAC) of iron oxides is 0.56 m$^2$/g (Alfaro et al., 2004). For BC we assume a MAC ranging between 5.9 to 7.5 m$^2$/g based on the measured BC size distributions (see section 3.3 paragraph 5) and those commonly reported in the literature (e.g., Bond et al., 2013), respectively. We calculated the relative absorption by Fe versus dust assuming that a range between 20-65% of the iron is light absorbing (this is a wider range than reported by Fairbridge to capture the full range of potential Fe that is light absorbing), and calculated the difference if the iron was made up of hematite (Fe$_2$O$_3$) versus goethite (Fe HO$_2$) (Lafon et al., 2006 shows that for some Chinese dust the fraction of iron oxides that is goethite and hematite are $\sim$73% and $\sim$27% respectively).

We scale the Fe mass to the ratio of hematite (Fe$_2$O$_3$) and goethite (Fe HO$_2$): Hematite conversion: ((56*2)+(16*3))/112 = 1.4 Goethite conversion: ((56+1+(16*2))/56=1.6 We...
on existing knowledge, these scenarios bracket the plausible range of dust absorption/BC absorption for the snow samples (n=70) collected from Mera Glacier. We also conducted similar analysis applying the MAC for Asian dust generically measured by Clarke (2004) (MAC=0.009 m²/g at 550 nm) and Yang (2009) (MAC=0.037 at 520 nm) to the gravimetric dust samples. Using a MAC for BC of 7.5 m²/g and the measured BC concentrations results in dust being 10-230 x more absorptive than BC based on the Clarke dust MAC and 40-937 x more absorptive based on the Yang dust MAC. If we multiply the BC concentrations x 5 to account for BC losses, dust is calculated to be 2-45 x more absorptive than BC based on the Clarke dust MAC and 8-187 x more absorptive based on the Yang dust MAC.

These results suggest that in general dust is likely more absorptive than BC, particularly for samples with higher impurity loads. This validates the results presented in Figure 4 in the manuscript, however there are several sources of uncertainty: a. Particle size will affect the assumed MAC for both dust and BC. b. It is not known how much of the Fe or dust is light absorbing c. Uncertainties in mixing between BC, dust and other constituents d. As addressed in the original manuscript in section 3.3, we need more information on how impurities accumulate at the glacier surface over the course of the year. The data presented herein suggests that when there are large dust concentrations dust dominates absorption, but when impurity loads are lower BC may be dominant. Considerably more work is needed to address the relative absorption of dust, BC and colored organics. There are too many uncertainties to conclude that absorption by BC deposited on snow/ice is trivial in the Himalayas. This study does, however provide important data suggesting that dust is playing a larger role than BC in absorption.

Moreover, many of the BC, Fe, and dust concentrations were determined in samples collected from crevasses, which may have modified the concentrations due to numerous processes occurring during or after the formation of the crevasse. These potential processes and their impact on the measured concentrations are not even mentioned in the manuscript, although the reported BC and dust concentrations in the snow are to my knowledge the highest ever reported from this region. Despite this additional uncertainty (which probably could have been avoided with the sampling in conventional snow pits), a large part of the conclusions regarding concentrations gradients and impacts are based on the concentrations from the crevasses.

- The results we present are the first observations from the south side of the Himalayas. A few months after this manuscript was submitted to ACPD, Ginot et al., 2013 submitted a manuscript to Cryosphere Discussions with dust and BC data from a shallow ice core from the accumulation zone of the same glacier. The reviewer is missing that these two studies provide the only observational BC and dust data from the south side of the Himalaya. We suspect that the largest reason there is a lack of observational data from this region is that the logistics of working in these areas are challenging (in the case of this study 5-7 days walk to access the glacier, and high elevation (6400 m a.s.l.) working conditions). The BC and dust concentrations from Mera La are high because this site is below the ELA of the glacier and more than one year of BC/dust deposition can coalesce into a single layer. The results presented in this manuscript provide highly needed data for understanding the role of impurities on glacier melt in this region. We have expanded the introduction to include the recent Ginot paper, and to stress the need of observations, particularly as they vary with elevation. While the results from Ginot et al. are a valuable contribution, their interpretation is problematic because they use accumulation zone BC and dust concentrations (which as we show are over a magnitude less than in the ablation zone) to estimate melt in the ablation zone of the glacier. Yasunari (2010) used atmospheric measurements to try to constrain BC deposition onto glaciers because of the lack of observational data. That there is a strong elevation gradient in BC and dust concentrations as we report is extremely important- the concentrations from the accumulation zone would minimally lower albedo, whereas lower down on the glacier impurity deposition is high and is most certainly accelerating melt. We address the reviewer's concerns about crevasse sampling following the reviewer's 'major comments' below. We did not dig snowpits at
Mera La because the firn/ice is too dense to allow this. While at Mera High Camp it would have been possible to dig a snowpit, we opted to sample the crevasse wall because considerably less exertion was required (which is not a negligible concern when working at these elevations), and by removing the outer surface of the crevasse wall we were able to sample the same snow as could be accessed with a snowpit.

In summary, the authors report some data and describe some methods, which would be extremely important for the community working on cryospheric sciences in this sensitive region. Unfortunately, they fall well short of a detailed description of all their methods (including their limitations) and the potential impacts of their results. The manuscript cannot be published in its current form. The reasons are described in more detailed in the major comments below. I support the idea of the anonymous reviewer # 1 that the data itself can be published in a journal like ESSD. However, only if the authors can demonstrate that the used methods are not seriously flawed.

Major comments: The sampling of snow in a crevasse adds substantially uncertainty regarding the snow stratigraphy and the measured concentrations. Fig. 3 shows that the top of the snowpack at the rim of the crevasse was variable and not well defined. The structure of the wall makes it apparently difficult to determine the exact depths for each sample even with a well defined snow surface. How did the authors determine the top of the snowpack and the depth for the samples? Although crevasse stratigraphy has been applied for more than 30 years to study surface mass balance of glaciers, its use has been restricted to high accumulations glaciers to minimize the disturbance of the stratigraphy. Moreover, crevasses form as a result of the movement of the glaciers and the resulting shear stress. They can be accompanied by liquid water formation. Did the authors investigate if such processes modified the observed stratigraphy? In addition, the polluted layers exposed in the horizontal wall may have undergone substantial sublimation. In the photograph in Fig. 3a cavities are clearly visible in the polluted layers possibly formed due to sublimation or even liquid water formation? It is difficult to see, but also the enriched layers in Fig. 3b seem to have different structure compared to the layers in between. Did the cavity formation and/or additional sublimation in the exposed crevasses contribute to an enrichment in BC and Fe in the samples and, thus, to the high measured concentrations? What was the aspect of the sampled crevasse walls? Nevertheless, sublimation possibly occurred throughout the exposed horizontal walls impacting not only the layer with high concentrations, but also other layers to a lesser degree. As a result, the measured background, average, and maximum concentrations could all be biased high in both crevasses even after the removal of 10 cm of snow. Therefore, the measured concentrations from the crevasses remain questionable and should be handled with caution. All these aspects regarding the sampling in the crevasses are not discussed in detail nor even mentioned in the manuscript.

Depths were determined by identifying the horizontal surface that marked the top of the profile, and using a measuring tape to measure depths along the sampling transect. In the crevasses sampled the impurity layers were oriented horizontally and visual inspection revealed minimal variability in the depths of the impurity layers spatially, as can be observed in 3a of the manuscript. The concerns of the reviewer would be more valid if we were using the stratigraphy in a mass balance study, but for the objectives of this study the depths of the impurity layers that we reported would vary little depending on where on the crevasse the samples had been taken. Because the depths of the impurity layers do not inform the interpretation of the data presented in the paper, the reviewer’s concern seems to have limited relevance. The crevasse walls sampled at both Mera La and Mera High Camp were northwest facing. This detail is now included in the manuscript in section 2.2. In the same section we further clarify how the crevasse was sampled: “Because the surface of crevasse walls can undergo sublimation and melt processes, the outer surface of the crevasse walls were removed using a mountain axe to create a fresh sampling surface, with a minimum of 10 cm of the fresh surface being removed.” Based on over a decade experience sampling snowpits and ice cores from mountain environments, I am confident that the sublimated surface was removed and the material sampled would be the same as what would be collected at
the same point on the glacier using a shallow ice drill. The uncertainty in
the measured concentrations also concerns the discussed altitude gradients, the
impact of the impurities on the albedo and the radiative forcing (Ch. 3.3), the implications
for the glaciers and the snowmelt (Ch. 3.4), and the conclusions (Ch. 4). An enrichment
of the impurities in the exposed crevasse walls may also be concluded from a
comparison with the snow pit data from November 2009. While in the summer lay-
ers of the snow pits sampled at the same locations (Mera La and Mera High Camp)
measured concentrations remained below 1 µg L⁻¹ (page 33501, line 6), the smallest
measured concentrations in the samples from the crevasse walls were 2 µg L⁻¹
with average background concentrations around 8 to 10 µg L⁻¹ (all values estimated
from Figs. 3a and b). However, at the only location where the samples originated in
both cases from snow pits, background concentrations were similar. Fig. 3c shows that
the measured BC concentration remained below 1 µg L⁻¹ at depth greater than 15 cm.
Higher concentrations were only determined in the surface layer, possibly also due to
sublimation?

- Higher concentrations were measured only in the surface at Mera Summit because
the snowpit wasn’t deep enough to encounter previous non-monsoon layers (winter-
spring).

The authors argue that the samples from November 2009 are less reliable due to the
long storage at ambient temperature (which was certainly not helpful) (page 33496,
lines 7f)? Nevertheless, I find the differences between the snow pit and crevasse sam-
ple striking, but unexplained in the manuscript.

- Examining the data again, we find the following: Background BC at Mera High camp
from the spring 2009 measurements was 2 ug/L, whereas it was 1 ug/L in the samples
from November 2009. Peak signal=318 ug/L

At Mera La, background from the spring 2009 measurements was 2-10 ug/L, whereas
from November 2009 the concentrations were ≤1 or less, with the exception of two
samples that measured 3 and 7 ug/L (this information is updated in the manuscript.
Peak signal=3535 ug/L

Because of the longer time that the November 2009 samples sat in storage, detailed
analysis on these small differences isn’t warranted and doesn’t indicate enrichment in
the crevasse profile. The result that is of interest is the effect that these impurities
are having on albedo and melt, and the high impurity layers as opposed to layers with
background concentrations are what are of relevance.

If the reported dust concentrations are correct, dust is the dominating factor for the
snow albedo and for further implications regarding snow and ice in the Himalayas (see
below). However, the description of the method of the determination of dust is limited
to two (almost identical) sentences (page 33496, lines 13ff and page 33503, lines
14ff) briefly indicating the limitations of the applied method, but without any further
specification of the uncertainties or errors. Throughout the manuscript only two values
are mentioned (both from the crevasse samples from Mera La). The authors definitely
need to expand the description of the applied method, the results, and potential errors.
If these values are correct, they would reverse our current understanding of the role
of absorbing impurities in the Himalayas. However, all other values on dust in snow
and ice in the Himalayas I am aware of are orders of magnitude lower.

- We have already addressed these comments in response to reviewer 2 above (see
detailed analysis on the gravimetric dust and use of Fe as a dust proxy to estimate ab-
sorption from dust versus BC). Fe concentrations at Mera High Camp and Mera Summit
are in the same range as those measured in the Everest ice core (Kaspari et al., 2011).
The Fe concentrations and gravimetric dust concentrations are considerably higher at
Mera La because of the elevational gradient and post depositional enrichment. This
highlights the need for the observational data presented in this study that documents the
elevation gradient.

The authors clearly state that the determined and reported BC concentrations are un-
certain due to two major effects: loss of particles during the process of transferring the snow into the gas phase using a nebulizer and due to the handling of the snow samples (melting prior to the analysis and long storage times). During their description, the authors refer several times to a publication in preparation by Wendl et al. suggesting that further details and tests are, or will be, described in detail in this manuscript. This technical manuscript should be published (or at least submitted) first.

-This manuscript has been accepted by Atmospheric Measurement Techniques Discussion: Wendl, I., Menking, J. A., Farber, Gysel, M., Kaspari, S., Laborde, M., and Schwikowski, M., Optimized method for black carbon analysis in ice and snow, Atmosphere Measurement Techniques, 7, 3075-3111, 2014.

The authors claim that they determined an altitude gradient in the measured BC gradients. While I understand the arguments for the gradient regarding the maximum concentrations mainly due to dry depositions and post-depositional processes, this is much less obvious for a similar gradient regarding the background concentrations. These are probably mostly influenced by wet deposition according to large-scale precipitation events during the monsoon period. Isn’t it more likely that such events lead to rather homogeneous concentrations for an altitude range from 5400 to 6400 m a.s.l. Even post-depositional effects impacting the concentrations in the surface layers are limited due to the high accumulation during a relatively short period.

-To address the elevation gradient in summer monsoon snowfall would require measuring BC concentrations in fresh precipitation. Such a data set doesn’t exist (logistics of working in this region during the summer monsoon are more difficult than the dry season). One prior study by Kang et al. (2001; Elevation Distribution of Precipitation Chemistry in Middle/Upper Troposphere in Summer, the North Slope of Central Himalayas, Environmental Science, 22, 2) measured major ions in fresh snowfall on Dasuopu glacier between 5800-7000 m a.s.l. during June 1997, and found that concentrations of calcium, magnesium and nitrate decreased with increasing elevation, whereas sulfate increased. The available atmospheric data indicates that atmospheric loadings of BC and dust decrease with elevation (e.g., concentrations at NCO-P are lower than in the surrounding lower elevation areas where atmospheric data is available). While the available data are limited, they do suggest that BC concentrations in snowfall are not homogenous with elevation.

The authors suggest that the albedo in the visible range was different at different elevations on the Mera glacier (page 33502, lines 5ff). This statement is based on the photograph shown in Fig. 2b. While the albedo may have been different, I am not convinced that such a statement can be supported by a photograph. The apparent albedo on a photograph depends on many parameters like the incoming radiation, the solar angle relative to the aspect of the terrain and the location of the camera, the settings of the lens and aperture, and atmospheric parameters. For example, in Fig. 2b the albedo in the region Mera La seems to be very low, while the apparent albedo at the surface in Fig. 3a also for Mera La seems to be very high.

-The reviewer is correct that the photograph can’t be used to support this statement. We’ve omitted the short paragraph where this was stated (end of section 3.2).

The authors state that the maximum observed concentrations at 154 cm depth at Meraï£ijLa may “represent convergence of multiple years of impurities” (Page 33503, line 23). This statement is in contradiction to the conclusion presented in ch. 3.1 that higher concentrations correspond to the winter-spring layer and the thick low concentration layers can be attributed to the snow from the summer monsoon (Page 33501, lines 1ff).

-In response to Referee 1’s point #3 and the results presented in Wagnon et al., (2013), we had already modified our interpretation of the data at the end of section 3.1, and now address possible additional causes of the low concentration snow observed at Mera La.

The calculations of the radiative forcing are based on the albedo values calculated with the SNICAR model. Unfortunately, numerous parameters to reproduce the calculated
values are missing.

-The reviewer points out that we did not include all of the parameters that were used to constraining the modeling of spectral and broadband albedos.

I was able to simulate the average and spectral albedo for the pure snow and the snow containing 258 µg L-1 BC with the online version of the model (snow.engin.umich.edu) using for example the default parameters for the snow thickness (1 m) and density (0.2 kg m-3). Are these the parameters used by the authors?

- No, these are not the complete set of parameters that we used. We provide them below.

However, such a thickness is completely unrealistic because Fig. 3a shows that the assumed BC concentration was only encountered in the uppermost sample maybe representing the top 3 cm. Below this layer the measured BC quickly drops to values around 100 and 50 µg L-1. Such a thin snow layer with high concentrations of impurities has obviously a much smaller impact on the albedo compared to a 1 m thick layer. As a result the presented albedo values may be greatly underestimated compared to realistic calculations using the observed profile. This effect is probably much smaller in the case of dust, which is an efficient absorber using the assumed concentrations. However, the decrease in Fe and, thus, in Fe is even more pronounced. By the way, the size range used for dust has an important impact in the SNICAR calculations. What size range was used? In summary, a full calculation of the albedo using the entire observed profiles is possible with the SNICAR model and needs to be performed to obtain reliable albedo values. Only with these values the instantaneous radiative forcing can be estimated.

- We do use them, they just were not listed in the manuscript. These are as follows: Grain radii – 350 and 750 µm Surface layer thickness – 0.02 m Lower layer thickness – 9.98 m Surface layer snow density – 500 kg/m3 Lower layer snow density – 350 kg/m3 Surface layer BC concentration – 258 ng/g Lower layer BC concentration – 5 ng/g Surface layer dust concentration (when used) – 9.3 mg/g Dust size distribution – the SNICAR model's 4 bins that cover the size distribution in Painter et al [2007], Lawrence et al [2010].

Finally, the authors present calculations for MAC values varied by less than 30 %, while the uncertainty in the measured BC concentrations (estimated to be larger than 60 % alone to the nebulizer and a possibly even larger uncertainty due to the sample storage, Ch. 2.2) and how they translate into uncertainties of the simulated albedo is not even mentioned here. The same is true for the uncertainty in the assumed dust concentrations.

- Actually, we do – but as you realize looking at the caption – the uncertainties in the MAC appear to be swept up by the vast majority of radiative forcing by dust. See below.

Ch. 3.4 does not bring substantial new information. First of all, many statements are related to the high measured BC concentrations in the crevasse walls, which are uncertain (see above). However, the further discussions here remain extremely general repeating already well known information: impact of absorbing impurities is largest in the 4000 to 6000 m altitude range because of the by far largest snow-covered area; absorbing impurities can have an impact on glacier mass balance, water resources, and radiative forcing; melting of snow and ice is accelerated by BC and dust; other absorbing impurities can also contribute, but the contributions of the different species remain difficult to quantify.

- We disagree that this section doesn’t provide new information. The first paragraph considers the role that the impurities play during the summer time, which is an area that further research needs to address, and is worthy of discussion in this manuscript. The second paragraph uses the first observations from the south side of the Himalaya to consider the north-south gradient in BC concentrations, which hasn’t been possible to address previously. The third paragraph addresses the elevation gradients in BC, and the relevance to snow covered areas.
Before. Prior studies on the Tibetan Plateau have looked at elevation gradients in BC at different sites (where varying distances from emission sources convolutes determining an elevation gradient), however the only other study to our knowledge that presented an elevation gradient of BC along a glacier was Wang (2012) in a methods paper, and this wasn’t put into context with snow extent. The remaining discussion puts the findings of this manuscript into the broader context of the discipline.

Instead, the following more important conclusions are warranted if the authors are convinced that their determined concentrations are correct. The overwhelmingly high dust concentrations dominate the snow albedo. Fig. 4 clearly shows the negligible impact of BC on the albedo in the presence of 9.3 g L⁻¹ dust even with the high BC concentrations reported here. (I am convinced that the negligible contribution of BC will not change with a correct calculation of the albedo as proposed above.) In that case, BC concentrations do not matter regarding all discussed further impacts on snow and glaciers and so on. According to their own calculations it is incorrect to write that “the impact of BC is diminished in the presence of dust” (page 33509, line12). With the proposed values for dust and BC, the impact of BC is negligible. In fact, it also does not really matter if the BC concentrations measured with the SP2 are correct or not. In contrast, the sources of dust (anthropogenic vs. natural?) and its behavior in the snow becomes more important and needs to be studied. Other absorbing impurities (brown carbon, organic compounds) will become only important (and need to be studied only) if they can compete with the high dust concentrations. The same applies to ch. 4.

-See our first response to Referee #2 where we provide detailed analysis on the relative absorption of dust versus BC, which is included in the revised manuscript. Based on the concentrations of dust, we are confident that at the time (April) these samples were collected, the radiative forcing by dust far outweighs that of BC. However (and this is a strong however), we have no idea how recently or frequently dust makes its incursions into the mountain snowpack. It is arguable that BC may be deposited on the mountain snowpack before dust events begin in the spring and in between dust events – given the proximity of BC sources to the mountains. In such a scenario, BC may drive warming and melting in the absence of large dust layers. We really do not know – we have suspicions, but in the absence of sustained measurements, we will not know. The measurements and modeling we present here are telling at that time of year and suggestive of what might happen in the rest of the ablation season of the spring. Note that this was already mentioned in the Conclusions of the paper, “However, the time span of the BC exposure at the snow surface in the dry winter-spring season is likely a persistent forcing before impurity convergence, but is not addressed by these single measurements.” Our analysis suggests that there may be times of the year when BC may dominate absorption, or when BC and dust absorption are comparable (see figure 2 of Fe concentrations vs. dust absorption/BC absorption). We agree with the reviewer that we can strengthen the conclusions that dust dominates albedo reduction, but considerable more work would be needed in order to make the broad conclusion that BC deposited on snow/ice is negligible in the Himalaya.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 33491, 2013.
Fig. 1. Fe (ng/g) measured via ICPMS vs. Gravimetric solids (g/L)

\[ y = 8\times10^{-5}x + 4.7161 \]
\[ R^2 = 0.85627 \]

Fig. 2. Fe (ng/g) vs Dust Absorption/BC Absorption