

The authors would like to thank the anonymous reviewers for their thorough reviews. We feel that the manuscript is significantly improved due to the reviewers' comments. Most notably, we have included substantial additional discussion on the under-catch issue as well as the surface vs sub-surface comparison (including an additional figure). Each of the reviewers' comments are addressed individually below. The reviewer comments are in black and our responses are in red. A copy of the manuscript with changes highlighted using "track changes" is attached to the end of the reply to reviewers.

Reviewer 1:

(1) The significant defect in this paper is a lack of discussion of the vertical variation of particulate concentration in the top few centimeters. At each site the authors sampled both surface snow (top 2.5 cm) and subsurface snow, as indicated on page 5083 lines 25-30, but the paper does not show results for the difference between the two levels. Melting and sublimation are expected to enhance the surface concentrations, as the authors note, and it is the top 2.5 cm that are most important for albedo. The authors did observe huge differences between summer and winter layers in one crevasse wall, but did not discuss the results of their paired surface-subsurface samples from their geographical survey. For example, page 5089 line 19-20 states that for Figure 7 the surface and subsurface samples were averaged; why was this done?

The dry season in the Cordillera Blanca is not without precipitation. Typically, about once per week or so in the dry season, there is precipitation generally amounting to a few to 20 centimeters of new snow. This leads to unexpected results at times when comparing the surface snow to the sub-surface snow. The surface snow sample is expected to contain more light absorbing particles as the reviewer states, due to accumulation at the surface from melting, sublimation, and dry deposition. In numerous cases during our sampling, snow was sampled immediately after a fresh snowfall. If the snow event resulted in only a few centimeters, the surface sample would be composed solely of snow from the recent snow storm while the sub-surface might contain older snow and thus snow from the previous surface. For this reason, often the sub-surface sample contained more light absorbing particles than the surface sample. Upon reviewing the paired measurements, the difference in eBC between the surface and sub-surface varied by up to a factor of 10 with a similar number of pairs having higher concentrations in the surface sample as in the sub-surface sample. A new section and figure have been put into the manuscript to discuss this part of the work. The figure shows the frequency of occurrence of different ratios of surface vs subsurface values.

(2) Undercatch. The quartz fiber filters apparently did not capture small particles efficiently, as the authors indicate (p 5086 lines 5-7 and 24-29.). This observation is consistent with the thorough study by Torres et al. (2013) on the collection efficiency of quartz fiber filters, which should be cited. For typical BC concentrations in rainwater, Torres found a maximum recovery of 38%. (Aerosol Science and Technology, DOI:10.1080/02786826.2013.868596)

The fullerene soot calibration BC that was used in our tests was created with atmospheric sized BC particles which generally ranged from 0.2 to 0.8 microns. BC size distributions showed that the quartz filters were missing a high percentage of the smaller particles. For that reason, we started using the

Millipore 0.22 micron filters as well. In tests the Millipore 0.22 micron filters collected 97% of the mass and nearly all BC particles larger than .2 microns in the standard. The downside to the Millipore 0.22 micron filters is that it takes a lot longer and much more force to filter the samples rendering them nearly impossible to use in the field. In 2014, for approximately 20 measurements in the Cordillera Blanca, near Cusco, and in Bolivia, snowmelt was filtered through the quartz fiber filters, then collected and filtered through the 0.22 micron Millipore filters immediately thereafter. In the most extreme case, the eBC determined by the LAHM technique for the Millipore filter amounted to 20% of the eBC value measured on the quartz filter. The average was closer to 5% with there often being undetectable levels of particles on the 0.22 micron filters. These tests were conducted on samples with quartz filter eBC values ranging from less than 5 ng/g up to 100 ng/g (quartz filters) and no discernable trend was observed in the 0.22 micron eBC percentage with respect to the quartz filter eBC concentrations. As there is some dust in the samples, it is suspected that either the BC can stick to the dust particles and/or dust particles may substantially clog the pores of the quartz filters reducing the effective pore size sufficiently to enable more efficient capture. Also, this the Cordillera Blanca snow often went through at least one and perhaps several snow transformations before being collected which tends to lead to aggregation of the BC particles.

(3) The calibration curve. Page 5086 line 16-18. The reason for the nonlinearity in Figure 3c is that at high loading the incident light is absorbed by the topmost particles, so the lower particles will not be exposed to much light. The curvature in Figure 3c indicates that the method will be unreliable for loadings >30 micrograms. In Figure 5, the outlier at rBC=75 is beyond the end of the calibration curve in Figure 3c. Perhaps this point could be used to extend the calibration curve.

This is a very interesting idea that is quite likely to be part of the non-linearity. For further measurements, we have adopted the tactic of reducing the amount of water filtered. Also, 30 micrograms on the filter does not equate to 30 ng/g as filter amounts are often reduced when the filter gets particularly dirty quickly. As we have gained experience, filters are now loaded until they are approximately 50% gray or until 600 ml (10 syringes) has been filtered. The volume of water filtered is always noted so that the collected particles can be normalized by volume of water. The rBC=75 measurement was at a location where only 300 ml of water was filtered, so the actual loading wasn't substantially higher than the calibration filters. This possibility the top layer of particles absorbing more than hidden layers below is now noted in the text.

Abstract Line 17. How were the samples kept frozen during transport?

The 2013 samples were kept frozen in an ice chest within another ice chest. The entire package was placed into a freezer when available (after 8 hours on a bus and again after 12 hours of aircraft flights). The majority of the samples were pristine upon arrival in Colorado and a few appeared to have been moderately transformed. The samples that appeared to have been severely transformed or completely melted were not included in the results of this publication. Since 2013, it has been found that high quality thermoses will keep snow frozen for several days without the need for re-cooling during transport. The trick appears to be to pack the sample vials in snow, then freeze the entire thermos in a freezer that is turned quite low. Place the thermos in a sleeping bag when packed for travel.

Page 5083 lines 8-9. Warren and Wiscombe 1980 is the wrong reference here. Cite instead Doherty et al. 2010 ACP.

The reference has been changed.

Page 5084 lines 4-5. How long did the melting take? BC may adhere to the wall of the Ziploc bag if meltwater is in contact with the plastic for a long time.

Melting generally takes 5-10 minutes, and filtering is done immediately after melting. From snow to filtering being completed is generally less than 20 minutes and volunteers are instructed that the filtering needs to be done as soon as the snow is melted. The stated time in the text is 20 minutes. The text has been clarified that this is from the start of melting to the finishing of filtering.

Page 5086 line 13. "32.5 micrograms". The points in Figure 3 are plotted at 30 not 32.5.

It was determined after the original writing of the manuscript that the 32.5 microgram filter may have been damaged and have let some BC by the edge of the filter. We eliminated it from the calculation. We have adjusted the text to refer only to the filters used.

Page 5087 line 15. A better reference is Grenfell et al. 2011 Applied Optics, which carefully defines "effective black carbon".

The reference has been changed.

Page 5090 line 2. How far is Huaraz from Regions 3,4,5?

Distances from Huaraz now shown in this paragraph.

Figure 3c. Exchange the x and y axes, because temperature-increase is the dependent variable.

This change to the figure has been made.

Figure 5 should be a square not a rectangle, since the same units are on both axes.

Changed

Figure 5 horizontal axis. Define "rBC".

A reference is now given for rBC in the text and it is spelled out on the axis and in the caption.

Figure 6. Exchange the x and y axes, because altitude is the independent variable.

It is common convention in atmospheric and cryospheric sciences to plot altitude on the y-axis. We prefer this convention when dependent variables are plotted versus altitude.

Figure 7a. Add a latitude-longitude grid.

Latitude longitude grid added.

Figure 7b. An alternative way to plot these data would be to use the distance from Huaraz (km) as the x-axis.

Each of the valleys are unique and in some valleys, numerous mountains are climbed and sampled. It is thought that the valleys are the main drivers for the local meteorology and pollution transport, so it seems more reasonable to plot the data by valley. The distances between Huaraz and a central location in each valley (or the summit in the case of Vallunaraju) are now indicated in the text.

Figure 7 caption line 5. Change “5 ng/g” to “8 ng/g”.

Caption changed as suggested.

Figure 8. Exchange the x and y axes.

As with figure 6, we prefer altitude as the y-axis.

Figure 9 vertical axis. The depth could be given in meters instead of centimeters; this would simplify the numbers.

Changed

Grammar and spelling

The title and abstract indicate that the topic is “particulates”; but page 5082 line 23 instead says “particles”. Are you making a distinction? If not, change all occurrences of “particulates” to “particles”, for simplicity and consistency. But “particulate” does still have a use; it is the adjective corresponding to the noun “particle”.

I agree and have changed all occurrences to “particle” or “particles”.

Page 5084 line 23. Change understand to understanding.

done

Page 5085 line 4. Change affect to effect.

done

Page 5087 line 3. Change suggest to suggests.

done

Page 5089 line 13. Delete “to”.

done

Reviewer 2:

Vertical variation: As Referee #2, I don't fully understand how the data from the surface and sub-surface samples are used. Are they all mixed together in the results, or are only surface samples used in some of the data presented? As Referee #2, I expect a discussion of the variability between these samples. You could potentially differentiate between these two types of samples in Figure 6.

As discussed in response to reviewer 1's question, there was a lot of variability which was determined to be caused by fresh snowfall. This led to the surface and sub-surface samples being up to ~10x different in eBC determined values for samples taken in the same location with no apparent preference as to which was higher. As there was no clear trend (on the average, surface samples had ~20% higher eBC values, but the standard deviation was substantially higher) it seemed reasonable to mix the results together. Please see the response to reviewer 1 for the changes made to the manuscript.

Undercatch: As Referee #1, I worry about the undercatch in the quartz filters. On page 5086 lines 4-6 the formulation is a bit peculiar for me. You use a BC standard that is not well captured by the quartz filters. This sounds a bit strange for me. Have you tested a BC standard with larger particles, similar to those that likely occur in the snowpack?

The response to reviewer 1 explains why we are confident that the undercatch is not a major problem. Regarding lines 4-6 on 5086, since it was determined that the quartz filters did not adequately capture all of the BC particles in the standard, the Millipore filters (which captured between 97 and 99% of the mass) were used to create the calibration filters. Thus, the LAHM instrument was calibrated with filters where the mass was accurately known with a possible undercatch of 3% of mass with the Millipore filters. It was determined both filter types (Millipore and quartz filters) behaved identically when clean in the LAHM system. The signal (temperature increase) for clean filters of both types was about 0.6C while a signal from 5 micrograms of BC was ~3.5C. This gave us confidence that the LAHM technique could detect levels of BC of less than 5 micrograms on a filter. As explained in response to reviewer 1, in tests conducted in 2014, a maximum of 20% undercatch was estimated with values generally averaging 5%. While we have made no claim that the quartz filters capture all of the snowborne BC we are confident that the quartz filters do capture a high percentage and this is also part of the reason that we report an effective BC value and state that it is possibly underestimated. The fact that the eBC values from the LAHM technique are well correlated with the rBC values from the SP2 and the further measurements described in the response to reviewer 1 using the two different types of filters gives us confidence that undercatch isn't substantially affecting our measurements.

Page 5081, line 25 to page 5082, line 4: References to BC emission statistics would strengthen your statements here. One candidate is Bond et al. (2004): A technology based global inventory of black and organic carbon emissions from combustion. Newer publications and more specific on the area around the Cordillera Blanca would be even better.

At this point, there are no black carbon inventories specifically developed for the region. Amazonian burning is said to account for about 50% of the carbonaceous aerosols in the amazon region (references

now in the manuscript). Coal is not commonly burned in South America, as hydroelectric power is one of the largest sources of electricity. More detailed inventories of emissions are currently not available.

Further, how large is Huaraz in terms of population size and at what altitude?

The population of Huaraz is about 100,000. Huaraz is at 3052 meters in altitude.

Page 5083, lines 10-13: Snow can often have a highly variable concentration of dust from nearby outcrops or other landscape features that act as sources of dust. From personal experience of sampling snow, variability in dust concentrations in snow is often observed visually at site. Could you potentially add a sentence on how you selected areas to avoid areas highly influenced by local dust and sand particles?

The Cordillera Blanca is no exception to dust from local sources. Volunteers are instructed to collect snow in areas at least 100 meters distant from visible rock outcroppings and to avoid areas where avalanches or avalanche debris could have contaminated the area with dust. Generally, the mountains are sampled by the easiest climbing routes which are less likely to be affected by avalanche. This is now explained in the text.

Page 5083, line 26 to page 5084 line 1: Can you explain better why you think the subsurface sample contain snow from the most recent snowstorm? Perhaps a reference to glacier or snow dynamics in tropic glaciers is in place and would help your statement? As the sampling is done in the dry season, I'm not totally convinced that the sub-surface samples contain snow from the latest snow storm. Could the snow come from different episodes and also include dry deposition and accumulation through melting?

Experimental ideas often don't pan out in practice, which is why I used the wording "should" for these statements. As discussed in the response to the surface samples compared to the sub-surface samples, your concern is obviously correct. Only after more substantial snow storms were the "previous" surface layers completely buried. That said, all measurements were collected at altitudes above the zone of ablation and volunteers were instructed to avoid any obviously old snow (that would be in the ablation zone). In the three expeditions (and 2014 as well), visually, seasonal snow was still present on glaciers down to 5000 meters or lower. This is addressed in the new section on the difference between the surface and sub-surface measurements.

Page 5087, lines 23-24 and the following sentences: The statement about the correlation between dust content and heat capacity is not very firm. Could you find any support for this finding in the literature? I understand this is not the main scope of the publication, but a strengthening of this part would be helpful.

This statement was made based on results from filters collected in other regions which contained very high concentrations of dust that were subsequently analyzed using the LAHM technique. The dusty filters were collected after a dust storm had very high eBC values (50-100) and by color were very different from Cordillera Blanca filters. The temperature curves measured by the LAHM instrumentation

for the dusty filters were substantially steeper than the steeper curve shown in figure 4. This discussion is now included in the manuscript.

Page 5088, lines 6-7: What is your reason to believe the reduction is 25%? I understand this issue concerns only a small part of the total samples, but I would like some more reasoning.

The filters that were affected had a pattern of concentric circles where the filter holder had been in contact. Upon removal, the particles that would have been on the filter were attached to the filter holder's concentric rings. The area where particles were removed was relatively easy to determine. 25% is a subjective estimate based on observation of the filters and the filter holder as well as the coloration of the "clean" circles on the filters. While not exact, it seemed better to adjust the values within the expected margin of uncertainty rather than average in values that were known to be incorrect. The values reported in the figure were only increased by 5 to 10% when the 25% was added to the affected filters.

Page 5088, lines 15-17: Could you please clarify whether the snow was tested by both LAHM and SP2, or if snow samples from the same sites were used for the two measurements. Variability on micro scale in the snow pack is well known; hence, it is very unlikely to get a perfect correlation if different snow samples from the very same sites are analyzed.

The LAHM filtering takes place in the field and it is therefore not possible to preserve the exact same water in a way that it would be useable for the SP2. Yes, it is very possible that some of the differences between LAHM and the SP2 measurements were due to micro-scale variability. Also, LAHM filtering uses on the order of one liter of water while SP2 measurements can be conducted with only a few grams of snowmelt.

Page 5089, lines 1-7: You could also compare to other regions, such as sites in Europe similar to that in your study.

More comparisons are now made in the text.

Page 5090, lines 16-17: How do you know the crevasse was newly opened? You don't need to elaborate about this in the paper, but I ask to check the quality in your findings.

I have personally climbed Vallunaraju every year for the past four years. It is relatively easy to identify how long crevasses have been open based on the amount of icicles on the more shaded side. There were very few icicles on the shaded side of this crevasse while in a crevasse a few hundred meters higher with the same aspect, there were numerous. Also, these samples were collected in 2012 and I did not recall there being a crevasse in that location in 2011.

Page 5090, lines 21-23: If possible, you should find and refer to literature on the stratification of annual layers in tropical glaciers to find support for your statement.

I asked numerous local glacier experts about these layers and this statement is made based on their responses. A technician who has been conducting glacier measurements for the Peruvian ministry of water for decades climbed Vallunaraju with me in 2014 and agreed with this conclusion.

Figure 3, caption line 3: Rewrite “of data 12 temperature profiles”. The current formulation is odd. x
Changed to “standard deviation for 12 temperature profiles shown in 3a”

