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Interactive comment on "Variability in individual particle structure and mixing states between the glacier snowpack and atmosphere interface in the northeast Tibetan Plateau" *by* Zhiwen Dong et al.

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Response to # Reviewer 2,

M. Dumont (Referee), marie.dumont@meteo.fr 2# Review of "Variability in individual particle structure and mixing states between glacier snowpack and atmosphere interface in the northeast Tibetan Plateau" by Dong et al.

Summary

This paper presents a dataset that explores the physical and chemical properties of light absorbing particles (LAPs) in the atmosphere and in the surface snowpack at several places in the Tibetan Plateau. Observations from TEM and EDW measurements are described. A tentative scheme to explain the observations is proposed along with an assessment of the changes in radiative impact.

Recommendations

This is a really interesting, rich and fascinating dataset, the conclusions drawn by the authors are of importance for a large community and may help reconciling current discrepancies between measured RF of LAP in snow and chemical content measurements. However the paper suffers from several flaws that need, in my opinion, to be corrected before the paper can be published as described in my specific comments below.

Author response: Thank you for all the positive comments and suggestions. We have revised the manuscript very carefully based on your review comments.

Specific comments

1/ My first major comment is that the data and methods description lack a lot of details that are essential for the reader to understand correctly the results and conclusion of the paper:

lines 84-101: Please provide more details on how the sampling was performed. The snow samples are taken at the same time of the atmospheric sampling? What is the volume? To which snowpack depth does it correspond?

Author response: we revised by providing more details of sampling, and glacier/snowpack surface samples were collected on the glacier/snowpack surface (with 5 cm snow depth, each sampled for 200 mL) for comparison with the atmospheric deposition process, and the snow samples are taken at the same time of the atmospheric aerosol sampling, see revised Line 104-123:

During the fieldwork sampling, we used the middle-volume-sampler (DKL-2 with a flow rate of 150 L/min) for TEM filter sampling in this study, with a flow rate of 1 L min−1 were used for TSP filter sampling in our study, by a single-stage cascade impactor with a 0.5 mm diameter jet nozzle and an airflow rate of 1.0 L min-1. Each sample was collected with 1 hour duration. After collection, sample was placed in a sealed dry plastic tube and stored in a desiccator at 25°C and 20±3% RH to minimize exposure to ambient air before analysis, and particle smaller than 0.5 mm can be collected efficiently by the instruments. In total, 80 aerosol samples were collected directly on the calcium-coated carbon (Ca-C) grid filter. Additionally, 88 glacier/snowpack surface samples were collected on the glacier/snowpack surface (with 5 cm snow depth, each sampled for 200 mL) for comparison with the deposition process, and the snow samples are taken at the same time of the atmospheric aerosol sampling. The detailed aerosol/snow sampling method is similar to the previous study in Dong et al. (2016, 2017). The information on sampling location, time period and aerosol/snow sample number are shown in Table 1. Snow samples were collected at different elevations along the glacier surfaces of the study. Pre-cleaned low-density polyethylene (LDPE) bottles (Thermo scientific), stainless steel shovel, and super-clean clothes were used for the glacier/snowpack surface-snow sample collection. All samples were kept frozen until they were transported to the lab for analysis.

Lines 102-114 : Though the measurements methods are described in some other references, it would be very useful to add here the main concept, uncertainties and limitations.

Author response: Yes, we have revised. See the revised manuscript Line 130-146:

The analyses involved conventional and high-resolution imaging using bright field mode, electron diffraction (Semeniuk et al., 2014; Li et al., 2014), and energy-dispersive X-ray spectrometry. A qualitative survey of grids was undertaken to assess the size and compositional range of particles and to select areas for more detailed quantitative work that was representative of the entire sample. This selection ensured that despite the small percentage of particles analyzed quantitatively, our results were consistent with the qualitative survey of the larger particle population on each grid.

Quantitative information on size, shape, composition, speciation, mixing state, and physical state was collected for a limited set of stable particles. Volatile particles, including nitrate, nitrite, and ammonium sulfate, though not stable under the electron beam, can be detected on EDX at low beam intensity. EDX spectra were collected for 15 s in order to minimize radiation exposure and potential beam damage. All stable particles with sizes 20 nm to 35 um were analyzed within representative grid mesh squares located near the center of the grid. Grid squares with moderate particle loadings were selected for study to preclude the possibility of overlap or aggregation of particles on the grid after sampling. The use of Ca-C grids resulted in clear and unprecedented physical and chemical information for the individual particle types.

Lines 115-124 : See comments 4

Author Response: Yes, we revised the issue. See the response to comments 4

Results and Discussion :

Whenever it's possible (description of Figures 3, 5, 7, 9 and 10) please quantify the mean and std differences between the snow samples and the atmospheric samples

Author response: Yes, thank you for the suggestion; we revised through quantifying the mean and SD differences between the snow samples and the atmospheric samples in the revised manuscript as shown below.

which indicates the LAPs composition in atmosphere of various locations as BC (mean percentage of 18.3%, standard deviation (SD) 2.58), OC (28.2%, SD 3.49), NaCl (11%, SD 2.58), Sulfate (17%, SD 3.49); Ammonium (4.8%, SD 3.01), Nitrate (7%, SD 2.83), Mineral dust (13.7%, SD 3.02), whereas the LAPs composition in glacier/snowpack surface as: BC (mean 21.3%, SD 2.49), OC (31.2%, SD 2.44), NaCl (16.2%, SD 3.12), Sulfate (6.8%, SD 1.32), Ammonium (2%, SD 0.81), Nitrate (3.3%, SD 0.95), Mineral dust (19.2%, SD 2.9). (See revised manuscript Line 205-211).

Figure 5 indicates that in atmosphere the composition ratio is as fresh BC (mean percentage of 29.7%, with SD 3.95), fresh OC (41.8%, 4.34), aged BC (9.8%, 4.02), and aged OC (18.7%, 4.11); while in the snow the composition ratio is as fresh BC (mean percentage of 8.4%, SD 2.71), fresh OC (17.7%, 4.42), aged BC (31.5%, 2.99), and aged OC (42.4%, 4.45). (See revised manuscript Line 259-263).

In Figure 7, the salt-coated particles in atmosphere accounted for mean ratio 54.61% (with SD 12.02) in various locations, while that in snow of glacier/snowpack was 18.59% (SD 7.04). (See revised manuscript Line 291-293).

As shown in Figure 9, the internally mixed particles of BC in atmosphere accounted for mean ratio 4.68% (with SD 3.07) in various locations, whereas that in snow of glacier/snowpack was 14.85% (SD 4.93). (See revised manuscript Line 305-308).

In Figure 10, the mixings states of BC/OC in the glacier/snowpack snow of northeast Tibetan Plateau showed that the internally, single and externally mixed BC/OC individual particles account for a mean ratio of 69.2% (22.5), 5,35(1.72), and 25.95% (with SD 22.4), respectively. (See revised manuscript Line 315-318).

Figures 2, 4 and 8: How was the classification performed? Please explain in the methods part.

Author Response: We add a supplementary material for explaining the classification of each kind of individual aerosol particles. Please also see Table S1 in the revised manuscript.

Classification criteria of sampled particle types, mixing states and their possible sources in the snow/atmosphere samples were indicated in Table S1. (See Line 203-204 in the revised manuscript)

Table S1 Classification criteria of sampled particle types, mixing states and their possible sources in the snow/atmosphere samples.

Line 161/162: Why are a-d representative of the atmosphere? And e-h of snow?

Author response: we have revised to make the sentence clear:

Figure 4a-4d is the representative particles of fresh BC/OM with fractal morphology and a large amount in the atmosphere, whereas Figure 4e-4h is the representative particles of aged BC/OM with aggregated spherical morphology in the glacier/snowpack surface. (See revised manuscript Line 240-243)

Lines 195-197: easily? Please explain how (in the methods part), and add a reference.

Author response: We revised to explain the reason for salt-coating easily observation in TEM, as:

Using TEM-EDX microscope measurements, we can also easily derive the salt-coating conditions based on the advantage of the transmission observation to obtain individual particle inside-structure (Li et al., 2014). Particle (e.g. BC, OM) with salt coating will appear clearly surrounded by various salts shell and with the BC/OM particle as core (see revised manuscript Line 146-150).

Thus we also delete the similar sentence in section 3.3.

2/ LAI is misleading (it also means Leaf Area Index). I would personally prefer the use of Light Absorbing Particles (LAPs) instead.

Author response: yes, good suggestion, we revised LAI to Light Absorbing Particles (LAPs) throughout the revised manuscript.

3/ The English is sometimes really difficult to understand and ambiguous. Though I am not a native English speaker, I would recommend a correction by a native English speaker.

Author response: yes, we have revised carefully throughout the manuscript, and also improved the English language by Elsevier language editing service.

4/ The RF change assessment is not detailed enough.

Line 118-124, please describe again the conditions and parameters used in the simulations. It is required here even if it has been described previously in another paper. Why were such contents selected for the simulation?

Author response: yes, we have revised. See revised Line 158-192 in the revised manuscript.

We also simulated the albedo change contributed by individual particle mixing states' variability of LAPs. The SNICAR model can be used to simulate the albedo of snowpack by the combination of the impurity of the contents (e.g., BC, dust and volcanic ash), snow effective grain size, and incident solar flux parameters (Flanner et al., 2007). In the SNICAR model, the effective grain sizes of snow were derived from the stratigraphy and ranged from 100 μm for fresh clean snow to 1500 μm for aged snow and granular ice. The model was run with low, central, and high grain size for each snow type to account for the uncertainties in the observed snow grain sizes. Snow density varied with crystal size, shape, and the degree of rimming. The snow density data used in the SNICAR model are summarized with low-, central-, and high-density scenarios for the model run based on a series observations in the TP and previous literature (Judson and Doesken, 2000; Sjögren et al., 2007; Zhang et al., 2018). In the model simulation, mineral dust (93.2$\pm$27.05 μg/g), BC (1517$\pm$626 μg/kg) and OC (974$\pm$197 μg/kg) average concentration data, as well as other parameters, such as effective grain size, snow density, solar zenith angle, and snow depth on the glaciers, are considered, and mass absorption cross-sections (MAC) for salt-coated BC is referred to the average situation derived from the northern Tibetan Plateau glaciers (Zhang et al., 2017, 2018; Yan et al., 2016; Wang et al., 2013). Though showing high level, the BC concentration data used in this study is comparable to the previous work results derived from ice core (Ming et al., 2008), with relatively much higher average elevation in Everest (its deposition site elevation 6500 m compared to 2900-4750 m a.s.l. of northeast Tibetan Plateau glacier sampling sites) and lower atmospheric BC concentration. While in this work we mainly focus on LAPs (BC, OC, mineral and others) in the glaciers and snow-

packs for the surface distributed impurities, which is often accumulated in summer with surface melting and with higher BC concentration.

When running the SNICAR model, BC was assumed to be coated or non-coated with sulfate (Flanner et al., 2007; Qu et al., 2014), or other salts. The mass absorption cross section (MAC) is an input parameter for the SNICAR model; it is commonly assumed to be 7.5 m2 /g at 550 nm for uncoated BC particles (Bond et al., 2013). For sulfate-coated BC particles, the MAC scaling factor was set to be 1 m2 /g, following Qu et al. (2014) and Wang et al. (2015). Other impurities (such as volcanic ash) were set to zero. In terms of the albedo calculation, RF due to BC and dust can be obtained by using Eq. (Kaspari et al., 2014; Yang et al., 2015):

Line 277-287, first describe the figure and the input for the different simulations. The difference in inputs is really difficult to guess

Author response: Yes, we have revised. See the revised last Results- section, revised manuscript Line 328-337.

Figure 12 showed the evaluation of snow albedo change of BC-salt coating change in the snowpack compared with that in the atmosphere using SNICAR model simulation in the MG, YG, QG, showing the albedo change of snow surface impurities in snowpack compared to that of the atmosphere. The parameters input for SNICAR model have been described in the method section. Mineral dust, BC and OC average concentration data, as well as other parameters, such as effective grain size, snow density, solar zenith angle, and snow depth on the glaciers, and MAC for BC were referred from the average situation in previous work of the northern Tibetan Plateau glaciers (Zhang et al., 2017, 2018; Yan et al., 2016; Wang et al., 2013).

Also see the revised Method section: Line 158-176, L185-196. We described the detailed input parameters for the model simulations for the LAPs in glacier snow.

We also simulated the albedo change contributed by individual particle mixing states' variability of LAPs. The SNICAR model can be used to simulate the albedo of snowpack by the combination of the impurity of the contents (e.g., BC, dust and volcanic ash), snow effective grain size, and incident solar flux parameters (Flanner et al., 2007). In this work, we use the online SNICAR model (http://snow.engin.umich.edu/). In the SNICAR model, the effective grain sizes of snow were derived from the stratigraphy and ranged from 100 μm for fresh clean snow to 1500 μm for aged snow and granular ice. The model was run with low, central, and high grain size for each snow type to account for the uncertainties in the observed snow grain sizes. Snow density varied with crystal size, shape, and the degree of rimming. The snow density data used in the SNICAR model are summarized with low-, central-, and high-density scenarios for the model run based on a series observations in the Tibetan Plateau and previous literature (Judson and Doesken, 2000; Sjögren et al., 2007; Zhang et al., 2018). In the model simulation, mineral dust (93.2$\pm$27.05 μg/g), BC (1517$\pm$626 μg/kg) and OC (974$\pm$197 μg/kg) average concentration data, as well as other parameters, such as effective grain size, snow density, solar zenith angle, and snow depth on the glaciers, were all considered; The mass absorption cross-sections (MAC) for salt-coated BC was referred to the average situation derived from the northern Tibetan Plateau glaciers (Zhang et al., 2017, 2018; Yan et al., 2016; Wang et al., 2013).

When running the SNICAR model, BC/OM was assumed to be coated or non-coated with sulfate (Flanner et al., 2007; Qu et al., 2014), or other salts. The mass absorption cross section (MAC) is an input parameter for the SNICAR model; it is commonly assumed to be 7.5 m2 /g at 550 nm for uncoated BC particles (Bond et al., 2013). For salt-coated BC particles, the MAC scaling factor was set to be 1 m2 /g, following Qu et al. (2014) and Wang et al. (2015). Other impurities (such as volcanic ash) were set to zero. In terms of the albedo calculation, the BC and dust radiative forcing (RF) can be obtained by using equation (1) (Kaspari et al., 2014; Yang et al., 2015):

5/ Overall, I think the methods and data part should be largely extended to describe in details all the methods used in the results part. In the results part, each Figure should

be described first and then discussed or commented. The reading is really confusing otherwise.

Author response: Yes, we have revised according to your suggestion. Also, see the detailed response to the above-related issues.

Minor comments

Table 1 – Some spaces are missing (Sampling dates column)

Author response: Yes, we revised. See revised Table 1.

Line 12 – “Aerosol impurities” this is quite redundant. Aerosols may be sufficient

Author response: Yes, we revised. “Aerosol impurities” to “aerosols”.

Line 14 - “significantly varied” → will cause significant changes in radiative forcing

Author response: Yes, we revised.

Everywhere: “glacier/snowpack” what do you exactly mean? The snowpack on the glacier? If yes, snowpack is probably sufficient.

Author response: Yes, we revised. We revise to “glacier and snowpack surfaces” here, as here we mean both the glacier surface snow and snow cover/snowpack samples in the high areas.

Line 110-114. I don’t understand this sentence. “Most” → please give a number.

Author response: Yes, we revised. It is writing mistake here.

Moreover, as the snow samples melting will affect the individual particle composition during the measurements, especially for various types of salts because the salt is unstable in high temperature (e.g. Ammonium and Nitrates) and will change, thus the snow/aerosol samples were directly observed under the TEM instrument and measured before it melted. All samples were measured in frozen states. (Line 152-157 in the revised manuscript)

Line 115: “evaluated” → simulated

Author response: Yes, we revised.

Line 160: into → onto

Author response: Yes, we revised.

Lines 186: between the interfaces → between the snow and the atmosphere

Author response: Yes, we revised.

Line 186-188: very complicated and ambiguous sentence.

Author response: Yes, we revised, as suggested by another reviewer the sentence is redundant with a similar meaning of the previous sentence. Thus we delete the sentence here.

Please also note the supplement to this comment:
https://www.the-cryosphere-discuss.net/tc-2018-166/tc-2018-166-AC2-supplement.pdf

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Interactive comment on The Cryosphere Discuss., https://doi.org/10.5194/tc-2018-166, 2018.