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The Cryosphere Discussions

Interactive comment on "Properties of black carbon and other insoluble light-absorbing particles in seasonal snow of northwest China" *by* Wei Pu et al.

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Response to Referee #1

We are very grateful for the referee's critical comments and suggestions, which have helped us improve the paper quality substantially. We have addressed all of the comments carefully as detailed below in our point-by-point responses. Our responses start with "R:".

Due to all of the formulas and special characters in our responses can't be added in text perform for the submission of interactive comments, we suggested that it should be better to look through the responses by the corresponding PDF files.

general comments: This paper deals with light absorbing impurities and their sources in snow in northern China. As the authors clearly state, these impurities impact upon the radiative budget of snow and are therefore important climate agents. These are relevant scientific questions within the scope of TC. This study applies the same general method used in several previous studies (Hegg et al., 2010; Wang et al., 2013; Zhang et al., 2013), and expands geographically on these studies, reaching similar conclusions as to the different sources responsible for those impurities in snow. The work is generally relevant to the scope of The Cryosphere and is worth publishing, once the authors take care of the following remarks.

R: Thanks for your comments. We have carefully responded the following remarks.

My major issues with the manuscript in its current form are:

(1) General lack of precision and clarity, making the reasoning of the authors very hard to follow.

R: We have revised the whole manuscript to make the manuscript more readable and comprehensible.

(2) Methodological problems with the PMF analysis: Recently, studies have shown the importance of uncertainties evaluation on the results of PMF on aerosols (ex: (Waked et al., 2014)). This lead to the publication of general guidelines for PMF analysis (Belis et al., 2014), that should be adapted here with more clarity. The authors refer to (in particular) (Hegg et al., 2010) for details on the PMF, but as they do not take the same species into account, there is a clear lack of details.

R: Based on the reviewer's comments, a new figure is given to reveal the uncertainty analysis as "Figure S1". We have also added more detailed description about the PMF analysis in section 3.4.1 in the revised manuscript as follow: The concentrations of the components (chemical and optical constituents) and the associated uncertainty datasets were used to run the PMF 5.0 model. General speaking, three to seven fac-

tors, twelve to thirty components and seven or more random seeds were applied with an objective step-by-step methodology in the PMF model to obtain the best solution in accordance with stability, accuracy, performance, and geochemical likeliness. This methodology is a multistep procedure. Firstly, all of the available components are included. Additional adjustments of the selected input components, numbers of factors and random seeds are dependent on an iterative process. We indicate that the choice of uncertainties could lead an important effect on PMF results. Then, we examined several tests for the uncertainties calculations which includes: (1) combining the detection limit (twice of the standard deviation of the blank samples) and the coefficient of variation (standard deviation of repeated analysis divided by mean value of the repeated analysis), which have been performed by Anttila et al. (1995) and Gianini et al. (2012); (2) the uncertainty datasets were calculated by considering the relative uncertainties of the concentration of each component (Waked et al., 2014). We also considered the results of the bootstraps and examinations of the residuals. The ability of the PMF model to replicate experimental concentrations, especially for components regarded as markers of the specific emission sources, is one of the primary principles applied to assess the permanence of the results at each step. More details in PMF model optimization can be found in Waked et al. (2014) and Belis et al. (2014). We used to estimate the fractional contributions to the 650-700-nm particulate absorption by all of the potential emission sources based on two reliable reasons: (1) represents the mass of BC, assuming all of the particulate light absorption (650-700 nm) is related to BC; (2) is only calculated based on the assumed MAC of BC; therefore, the errors of were the lowest among the studied variables. The chemical components were , , , , Na+, K+, , KBiosmoke, Al, Fe, Mn, Cu, Cr and Ba. Finally, a set of high uncertainty datasets was used in this study. For example, the relative uncertainty was 40% for , , and Al, 50 % for , and KBiosmoke, which was comparable with that used in other studies of the spatial variations (Hegg et al., 2009, 2010; Zhang et al., 2013a; Doherty et al., 2014). All these components were described from weak to strong in the PMF on account of their signal-to-noise ratio and the effect on tracing emission sources. The

results indicated that the , „ , Cu and Cr were categorized as “weak”. Therefore, the optimal number of factors/sources was four based on the robust and theoretical Q values (Hegg et al., 2009, 2010). However, three-factor provided more physically reasonable results and the most easily identifiable sources, which was consistent with studies of snow in northeastern China (Zhang et al., 2013a) and North America (Doherty et al., 2014). The diagnostic regression R2 value for with this three-factor solution was considerably high (0.87). Hence, we indicated that the three-factor solution was the best choice in this study.”

(3) In particular, the authors seem to be using in their PMF analysis some derived quantities such a Kbiosmoke. How does this concentration depend on assumption on seasalt and crust concentrations ratios ? And does it impact the PMF ?

R: The method to derive KBiosmoke concentrations has been described in section 2.2 and have been investigated in previous study (Hsu et al., 2009). For instance, the KBiosmoke as a well-known indicator plays an important role in tracing biomass burning emissions, which has already been widely used in the PMF analysis (Hegg et al., 2009, 2010; Zhang et al., 2013a; Zhang et al., 2013b).

(4) Uncertainties analysis: in general, I feel the authors should have done a more thorough uncertainty analysis. This is particularly true concerning the retrieval of absorption from iron oxide, Brown carbon and black carbon from the ISSW measurements. The authors mention (Doherty et al., 2010) and (Grenfell et al., 2011) for error estimates on those measurements, but these references only took into account Black and Brown Carbon, so only partially apply here. See for example the discussion by (Lack and Langridge, 2013)

R: Based on the reviewer’s comments, the following contents are added: “Lack and Langridge (2013) indicated that the attribution biases of BC absorption are from +7% to -22% by using the AAE in the range of 1.1 ± 0.3 instead of 1 as the common default. In order to reduce the uncertainty of BrC absorption at 404 nm less than $\pm 100\%$, the

absolute contribution from BrC absorption must be at least 23% (10%) of that from BC for PAS measurements. Significantly, the variation of AAE plays an important role in affecting the light absorption attribution. However, most of the studies only took BC and BrC (or OC) into account, which ignored the effect of mineral dust on light absorption. For instance, Doherty et al. (2010) revealed that the consideration of dust will add the complexity but does not effectively change the results due to negligible fractional light absorption of dust in some areas such as the Arctic. However, Wang et al. (2013) and Zhang et al. (2013a) indicated that the light absorption of mineral dust could not be negligible across northern China. Therefore, in view of the importance of mineral dust and the complexity of the combination of BC, OC and dust in snow, we did a sensitive test on two possible cases that the mixing ratios of BC, OC and Fe are 100 (15) ng g-1, 1000 (150) ng g-1 and 50 (50) ng g-1 with the fractional absorption of 42% (34%), 54% (43%) and 4% (23%) based on our filed measurements. We estimated the relative uncertainty of the attributed absorption assuming the AAEs of BC, OC and Fe are 1.1$\pm$0.3, 6$\pm$2 and 3$\pm$1 (Doherty et al., 2010; Lack and Langridge, 2013) instead of 1.1, 6 and 3. As shown in Figure S1, the relative uncertainties of BC and OC are from -53% to 29% and -25% to 43%, respectively, which mainly resulted from the variations of AAEs of OC and BC (left panel). The variation of AAEs of Fe has a slight effect on the light absorption. For Fe, the relative uncertainty is from -18% to 22% based on the variation of AAEs. In case 2 (right panel), the fractional absorption of Fe is much more important compared with that in case 1. The relative uncertainties of BC and OC increased and range from -65% to 35% and -40% to 61%, which highlight the varied AAEs of Fe in uncertainty analysis. The analysis indicates that the changes of AAEs of Fe on uncertainty estimates are dependent on the fractional absorption of Fe."

Specific Comments:

P3 L15: "radiative forcing is highly uncertain": did the authors mean radiative forcing in general, or more precisely in snowy places ? Please precise.

R: The sentence has been revised as "However, the regional and global radiative forc-

ing affected by the ILAPs in snow/ice is highly uncertain".

P5 L15: "dust is the main absorber in snow locations" : missing word, many ? Most ?

R: The sentence has been revised as "Recent studies have indicated that the light absorption by mineral dust is mainly related to iron oxides such as goethite and hematite (Alfaro et al., 2004; Lafon et al., 2004, 2006). Although its ability to reduce snow albedo is less than that of BC by approximately a factor of 50 (Warren, 1984). We note that the mass loading of mineral dust could be dominated in several snow sampling locations (Wang et al., 2013)."

P6 L1: "quantify the source attribution" $\rightarrow$ "attribute the sources"

R: "quantify the source attribution" has been revised as "attribute the sources".

P7 L2: "we evaluated the chemical components to examine the potential emission sources": quite vague

R: The sentence has been revised as "The chemical components and ILAPs were also used to estimated the potential emission sources and source attributions of ILAPs in seasonal snow."

P7 L20-21: how does measure the snow density and temperature help quantify the deposition flux of BC?

R: We are sorry for the misleading. The sentence has been rewritten as "Snow density and snow temperature were also measured within each layer, which could be useful for the parameterization of snow albedo modeling (Flanner et al., 2007, Wang et al., 2017). "

P8 L7: "nuclepore filters were subjected to BC and OC analyses": this sentence is overly misleading. BC analyses are optical measurements (see (Petzold et al., 2013) for nomenclature), which is actually what is done here. But OC generally refers to carbon measurements made from combustion methods, which is not the case here,

and would anyway be impossible on nuclepore filters.

R: We have revised the sentence as “the filters were used to measure the light absorption of ILAPs”. The details in separating the light absorption of ILAPs in snow could be found in Wang et al. (2013, 2017).

P12 L3-4: “quantify contributions from sources based on composition or fingerprints of the sources” : this seems ill-formulated as actually the PMF gives factors purely from statistical considerations, without any a priori knowledge of eventual “source fingerprints”. It is then up to the user to interpret the calculated statistical factors as sources, as the authors actually did

R: We have revised the sentence as “provides source attribution by identifying and quantifying source profiles and contributions prior, which is based on mathematical approaches”.

P12 L14-16: from this sentence, the choice of the number of factors seems pretty much to be an arbitrary decision from the user, whereas some “best practices” exist for this choice.

R: The sentence has been revised as “therefore, the number of factors is unknown priori, which must be selected individually in terms of stability, accuracy, performance, and geochemical likeliness of the PMF results and the analyst’s understanding of the sources”.

P14 L10-15: the authors mention a potential oulier. Is it the only one ? How were these accounted for in the PMF ?

R: Sorry for the misleading. If we only considered all of the values of during this snow field campaign, the bottom value of at site 83 should be considered as a potential oulier. Then, a possible explanation was given as “however, the underlying soil may have been responsible for this high value. Therefore, we note that this value should not be used to represent the regional background level of BC.”. We indicated that only the

chemical species and in surface snow were used as the input parameters for the PMF model. Therefore, the highest value of at the bottom layer at site 83 was irrelevant with the PMF analysis. Then, the following sentence is also revised as “After excluding the bottom value of at site 83”.

P20 L15-20: the authors mention “considerable errors”: could they be more specific ?

R: We plotted a new figure as Figure S1 to analyze the uncertainties in attributed absorption of BC, OC and Fe at 450 nm associated with the changes of Absorption Angstrom Exponent (AAE). More details could be found in comment (4) and Figure S1.

P21 L10-15: it would be good to compare the number of factors to the total number of species taken into account

R: We have compared the number of factors to the total number of components taken into account, and presented a more detailed description on PMF analysis. More details could be found in comment (2).

P21 L16: does really the Figure show “measured mass concentrations” ? Or is it rather calculated mass concentrations (calculated by the PMF)

R: We have revised “measured mass concentrations” as “calculated mass concentrations”.

P23-24: I do not really understand the interest of §3.4.2. As I understand, it discusses the contribution of a given source to each site, normalized by the average contribution. I do not really see what geochemical information this holds. On the opposite, I understand the following paragraph, where on each site, we have a picture of the origin of LAIs.

R: Based on the reviewer’s comment, we combine 3.4.2&3.4.3 as section 3.4.3 of source attribution in snow.

P25 L3-10: the authors point that their results differ largely (on the one common region)

with previous results from (Zhang et al., 2013), then invoke differences in species taken into account and inconsisntecies in the PMF analysis. This needs to be precised: if results vary too much upon the species taken into account, then there need to be a clear discussion no why your species set is “nest”

R: We have revised the sentence as “The PMF results in Qinghai in this study were not comparable well with those by Zhang et al. (2013a), who indicated that soil dust was the dominant source of ILAPs. However, the discrepancy could be concluded as (1) the receptor sites between two field campaigns were really far; (2) the chemical components inputs were different (e.g. and KBiosmoke instead of levoglucosan as the markers for biomass burning emissions); (3) the variables that characterized the particulate light absorption in the PMF analysis were inconsistent that we used instead of ILAPs to estimate the fractional contributions to the 650-700-nm particulate absorption.”. In addition, we have added a more detailed description on PMF analysis. More details could be found in comment (2).

P25 L15-20: the correlations showed in figure 9 do not seem very strong. Could the authors give some p-values for those ?

R: Based on the reviewer’s comments, the Figure 9 was replotted with the confidence level added (See Figure 9). As shown in Fig. 9a, the contributions from industrial pollution sources influenced by human activities is highly related to the altitude, while the biomass burning and soil dust only show weak correlations.

P26 L21: nitrate and sulfate are secondary aerosol, not primary

R: We have revised “primary” as “major”.

Fig. S1. Uncertainty in attributed absorption of BC, OC and Fe at 450 nm associated with the changes of Absorption Angstrom Exponent (AAE). Case 1 & 2 represent two typical conditions that the fractional absorption of Fe are 4% and 23% in seasonal snow during this field campaign, respectively.

Fig. 9. Scaled contributions from each source/factor as a function of altitude at sampling sites in Xinjiang. The contributions were normalized by the average value of the respective factor contribution over all sites.

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Please also note the supplement to this comment:
http://www.the-cryosphere-discuss.net/tc-2016-233/tc-2016-233-AC1-supplement.pdf

Interactive comment on The Cryosphere Discuss., doi:10.5194/tc-2016-233, 2016.

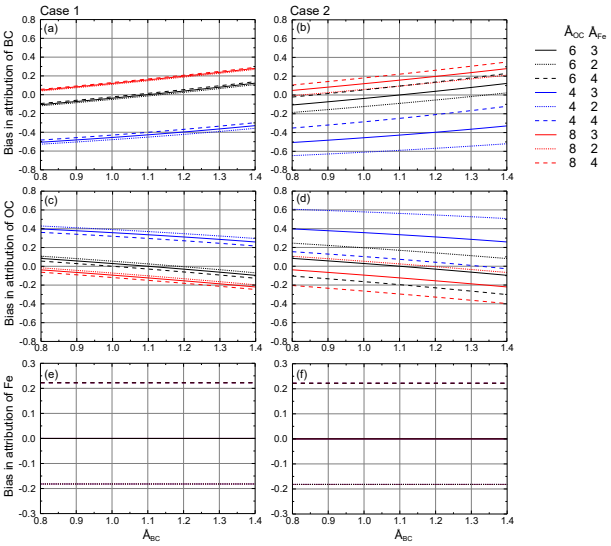

Fig. 1. Fig. S1. Uncertainty in attributed absorption of BC, OC and Fe at 450 nm associated with the changes of Absorption Angstrom Exponent (AAE).

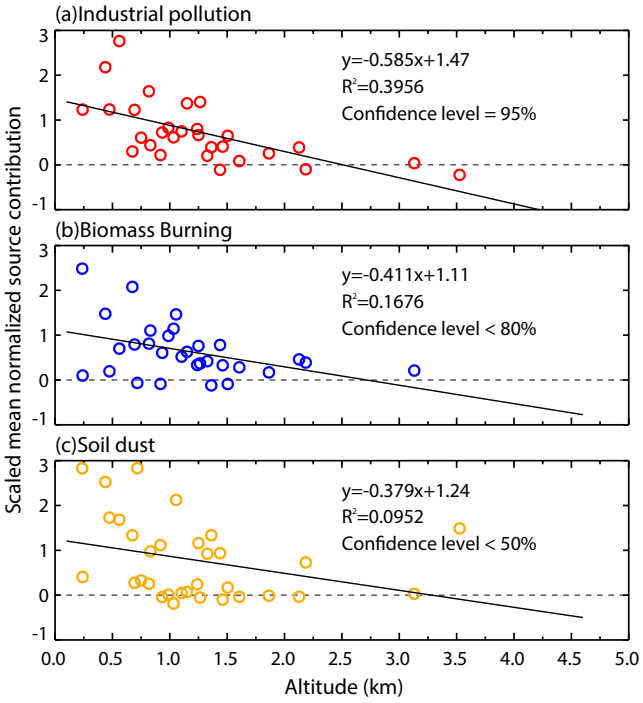

Fig. 2. Fig. 9. Scaled contributions from each source/factor as a function of altitude at sampling sites in Xinjiang.