

## 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:".

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 factors, 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  $C_{BC}^{max}$  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)  $C_{BC}^{max}$  represents the mass of BC, assuming all of the particulate light absorption (650-700 nm) is related to BC; (2)  $C_{BC}^{max}$  is only calculated based on the assumed MAC of BC; therefore, the errors of  $C_{BC}^{max}$  were the lowest among the studied variables. The chemical components were  $SO_4^{2-}$ ,  $NO_3^-$ ,  $Cl^-$ ,  $F^-$ ,  $Na^+$ ,  $K^+$ ,  $NH_4^+$ ,  $K_{Biosmoke}$ , 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  $C_{BC}^{max}$ ,  $Na^+$ ,  $K^+$  and Al, 50 % for  $SO_4^{2-}$ ,  $NO_3^-$  and  $K_{Biosmoke}$ , 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  $NO_3^-$ ,  $NH_4^+$ ,  $Cl^-$ ,  $F^-$ , 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  $R^2$  value for  $C_{BC}^{max}$  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  $K_{biosmoke}$ . How does this concentration depend on assumption on seasalt and crust concentrations ratios ? And does it impact the PMF ?

R: The method to derive  $K_{\text{Biosmoke}}$  concentrations has been described in section 2.2 and have been investigated in previous study (Hsu et al., 2009). For instance, the  $K_{\text{Biosmoke}}$  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  $1.1 \pm 0.3$  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)  $\text{ng g}^{-1}$ , 1000 (150)  $\text{ng g}^{-1}$  and 50 (50)  $\text{ng g}^{-1}$  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 forcing 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” → “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 outlier. 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  $C_{BC}^{est}$  during this snow field campaign, the bottom value of  $C_{BC}^{est}$  at site 83 should be considered as a potential outlier. 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  $C_{BC}^{max}$  in surface snow were used as the input parameters for the PMF model. Therefore, the highest value of  $C_{BC}^{est}$  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  $C_{BC}^{est}$  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 inconsistencies 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

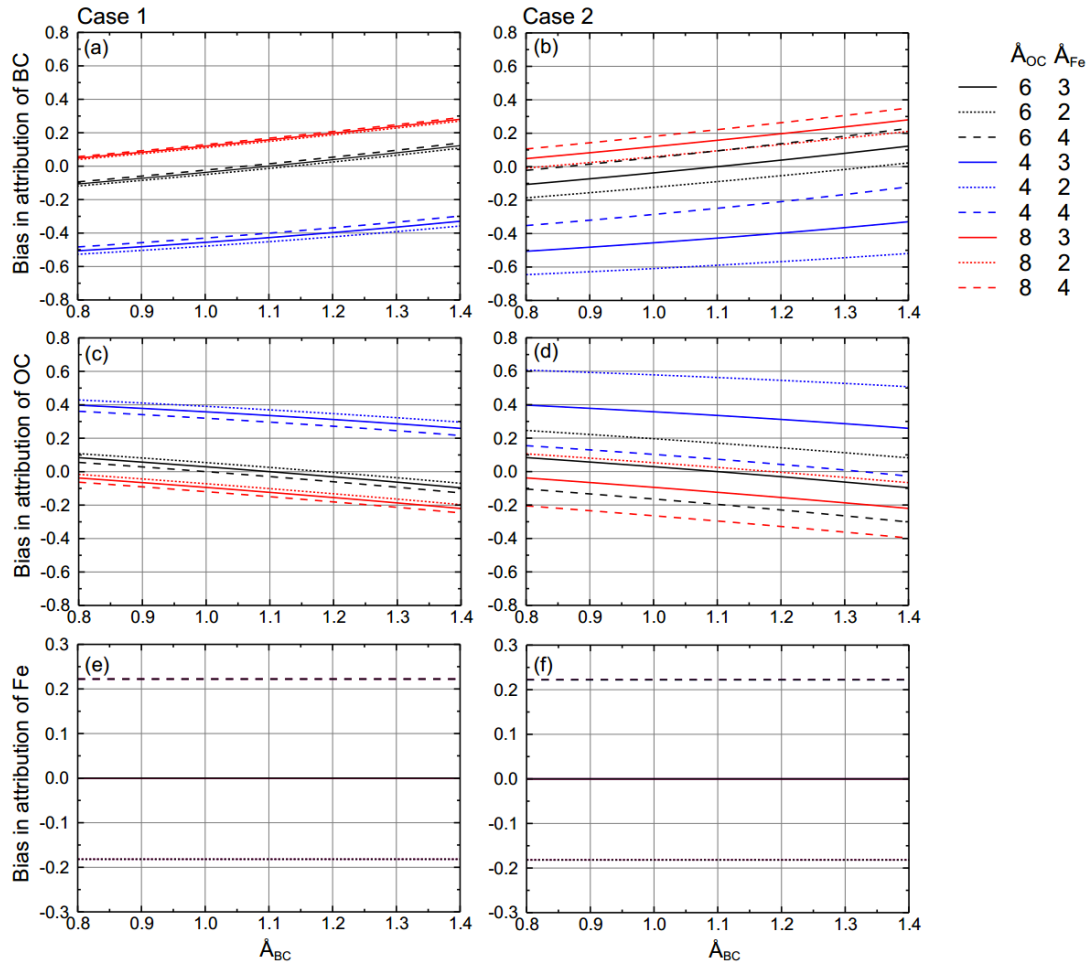
receptor sites between two field campaigns were really far; (2) the chemical components inputs were different (e.g.  $K^+$  and  $K_{\text{Biosmoke}}$  instead of levoglucosan as the markers for biomass burning emissions); (3) the variables that characterized the particulate light absorption in the PMF analysis were inconsistent that we used  $C_{\text{BC}}^{\text{max}}$  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.

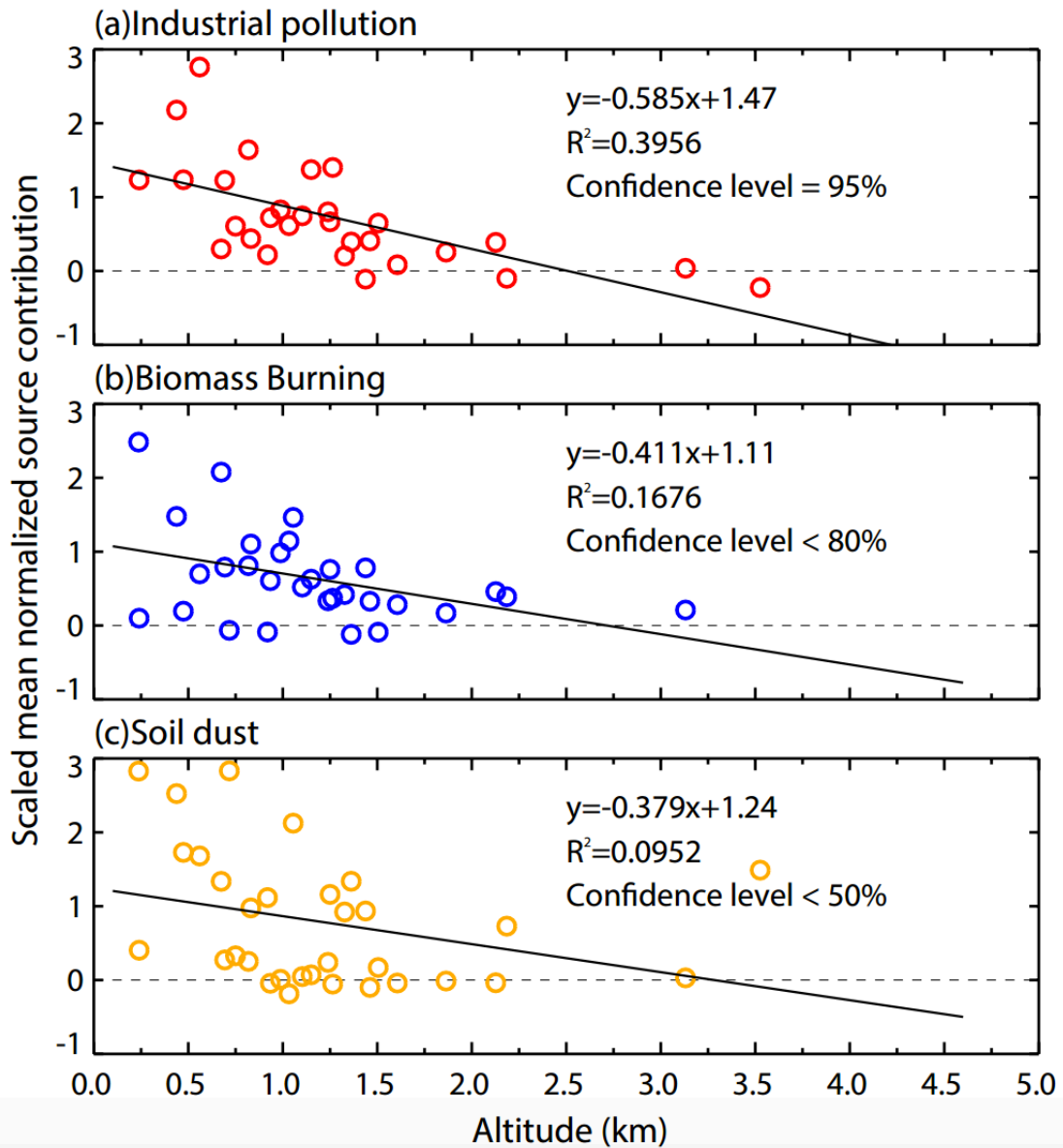
P26 L21: nitrate and sulfate are secondary aerosol, not primary

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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## Response to Referee #2

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:".

I have read the manuscript titled: Properties of black carbon and other insoluble light absorbing particles in seasonal snow of northwest China. Overall, I feel that the manuscript is well written and is worthy of publication in *The Cryosphere*. The first referee made multiple comments regarding the chemical analysis. As this is not my area of expertise, I will comment on some additional issues that I noticed.

R: Tanks very much for your comments and suggestions, we have addressed all of the comments carefully as detailed below.

This research (as well as many publications reporting on this topic) suffers from one common uncertainty. Since results are presented from data collected in one moment in time, how useful are the results in reality? How representative is that one point in time for representing conditions at any other time in the snow year? Different weather conditions can significantly affect the snow ILAP concentrations. While this is a common problem with this type of measurements that can only be overcome by more intensive sampling, I feel that the authors should include wording that state that the results are from one time measurements at each location and may not be representative of the long term characteristics of the snow in that location.

R: Generally, the sampling sites were selected 50 km away from cities and at least 1 km upwind of the approach road or railway to minimize the effect of pollution from local sources across northwestern China. But we also agree with the reviewer that understanding spatial and temporal differences of ILAPs in snow is still challenges. Therefore, the comparison of the seasonal and interannual variability of the ILAPs in snow was investigated, and the result shows that the differences of ILAPs in snow are relatively small in the Arctic and northeastern China (Doherty et al., 2010; Wang et al., 2013, 2017). We note that further snow field campaigns were still performed worldwide to limit the uncertainties of ILAPs in snow due to the spatial and temporal differences across northern China, the Himalayas, North America, Greenland and the Arctic since 1980s (Cong et al., 2015; Dang and Hegg, 2014; Doherty et al., 2010, 2014; Hegg et al., 2009, 2010; Huang et al., 2011; Xu et al., 2009, 2012; Zhao et al., 2014; Warren and Wiscombe, 1980, 1985). For instance, a similar paper on the mixing ratios of ILAPs in Arctic snow has been widely used for validating modeled snow BC mixing ratios (Doherty et al., 2010). Therefore, we indicated that the datasets in this study can contribute to advancing remote sensing techniques and reducing the uncertainties of the

model simulations to enhance our further understanding of the climate impacts of ILAPs in snow and ice.

If I recall correctly, the Hegg technique requires that the snow is relatively fresh.

R: We have carefully looked through the papers by Hegg et al. (2009, 2010). The major points in the literatures are the attribution of the chemical species and the mixing ratios of BC in seasonal snow, which were based on the results of light absorption of BC in both fresh and aged snow by Doherty et al. (2010). Recent studies also mentioned that using the chemical and optical data, which are included both fresh and aged snow samples, were input to a Positive Matrix Factorization (PMF) analysis of the sources of particulate light absorption (Doherty et al, 2014; Zhang et al., 2013). Therefore, the PMF technique is mainly based on different species as the input without considering the snow samples as fresh or aged.

There do not appear to be any comments regarding the time since the most recent snow storm for each of the sites (other than the mention of the 13 sites where it was snowing during collection). Could this affect some of the chemical analysis as some chemical constituents may have washed out of the snowpack?

R: We note that the snow field campaign was conducted in winter season and the surface snow kept frozen and hadn't yet melted during our sampling processes in most of the snow sampling sites. Therefore, the melting or washing effects were negligible in this study. But we also agree with the reviewer that the melting and washed out effect of the snowpack should be considered if the snow samples were really melted. Details for the melting and washed out processes of ILAPs in snow could be found by Wang et al., (2013, Equation 1 & Figure 3) and Doherty et al. (2013, Figure 1) .

Page 3 line 11, Using the SNICAR online model (<http://snow.engin.umich.edu>), using the default snow constants then either 0 or 10 ng/g, the broadband albedo reduction is closer to 0.3% rather than 1%. The Warren and Wiscombe (1980) paper shows possible values with some relatively extreme cases depicted. Could the authors clarify what snow conditions they are using and then clarify the appropriateness of these conditions?

R: We have revised the sentence as “Warren and Wiscombe (1980) indicated that a mixing ratio of 10 ng g<sup>-1</sup> of BC in snow with snow grain size of 1000 μm may reduce the snow albedo at 400 nm by approximately 1%.”

Page 14 line 19: Earlier the authors stated that they sampled in 5 cm steps down through the snow pack. Is this comparable to other studies where you compare the results?

R: Sure, the methods on snow collections and spectrophotometric analysis are definitely comparable with the previous studies (e.g. Doherty et al., 2010, 2014; Wang et al., 2013).

If the surface sample is the top 5 cm of snow, how does the density of the snow affect the measurement (if dry deposition on the surface is the main source, then the 5 cm of snow would dilute the measurement significantly with density as an additional factor).

R: We agree with the reviewer that it is still a challenge to separate the dry and wet deposition of the ILAPs in the top 5 cm snow. The most important reason is that the snow albedo is mainly influenced by the surface snow, especially for the top 5 cm. Therefore, in order to compare and improve the model simulation, we use the top 5 cm as the cumulative values, which include both wet and dry deposition. However, we also indicated the dirty layers and new fallen snow were collected in all sites separately, even for the filtration processes. Doherty et al. (2010) also indicated that if there was obvious layering, for example a thin top layer of newly fallen snow or drift snow, that layer was collected separately, however thin.

If all of the BC is on the surface, then sampling the top 1 cm of snow versus the top 5 cm of snow (assuming uniform density) would give you a factor of five difference in mass mixing ratio since BC is generally reported in a mass per volume unit.

R: The same as above comment. Generally, we collect the vertical profiles of snow sample in every 5 cm when the snow looks uniform. Otherwise, the snow samples will be gathered separately, if it is a significant dirty layer or new fallen snow, whatever 1 cm or thick layers.

Nomenclature: The different variable names are not well defined at their first use in the manuscript. Ie. What specifically do “MAX”, “EQUIV”, and “EST” in superscript mean next to C subscript BC? It might be nice to have these and the many others in a table for easy reference. If they are equivalent to something used in other literature (eBC in Grenfell), please list the equivalents as well.

R: Thanks for your suggestion. We have added the description of variables in Table 1.

**Table 1.** Variables derived by using the ISSW spectrophotometer.

Symbols	Description of variables
$C_{BC}^{max}$	Maximum BC is the mass of BC per mass of snow, if all particulate light absorption (650–700nm) is due to BC.
$C_{BC}^{est}$	Estimated BC is the estimated true mass of BC per mass of snow,

	derived by separating the spectrally resolved total light absorption.
$C_{BC}^{equiv}$	Equivalent BC is the amount of BC that would need to be present in the snow to account for the wavelength-integrated (300–750nm) total light absorption of down-welling solar radiation by all particulate constituents.
$\hat{A}_{tot}$	Absorption Ångström exponent is calculated between 450 and 600nm, for all particulate deposited on the filter.
$f_{nonBC}^{est}$	Fraction of light absorption by non-BC ILAPs, is the absorption by non-BC particulate constituents, weighted by the down-welling solar flux, then spectrally integrated from 300 to 750nm.

Page 18 line 2: Something is missing: “vertical differences were missing at could sites, . . .”

R: We have revised the sentence as “...although apparent vertical differences could be observed at could sites, such as site 47”.

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## Response to Editor

Dear Authors,

Both reviewers have an overall positive evaluation of your paper, but both also note significant deficiencies in your work, which may cast doubt on the robustness of your methods and conclusion. Reviewer 1 in particular is not fully convinced by your use of the PMF method. He also notes a lack of detail or erroneous points in your description of analytical methods. Reviewer 2 questions the time-representativity of your data set. He also makes an interesting comment regarding the radiative impact of BC in snow. I encourage you to submit a very carefully revised version, where all measurement uncertainties and data representativity are clearly detailed and their effects taken into account, in particular regarding their possible impact on the conclusions drawn from your PMF analysis. Your modifications will be evaluated critically before possible acceptance of your paper in The Cryosphere.

Best regards  
Florent Domine

We are very grateful for the editor's comments and suggestions, which are very helpful for us to improve and clarify the presentation of our results. The following are our key point responses to the editor's comments.

Reviewer 1 in particular is not fully convinced by your use of the PMF method. He also notes a lack of detail or erroneous points in your description of analytical methods.

R: We have added a more detailed description of the PMF method, which includes: (1) A more detailed analysis process; (2) The methods of uncertainties calculations; (3) The principles for selecting the optimal PMF results. We also presented a description of the input components and the values of the associated uncertainties in our study. In addition, the character of each component was identified according the signal-to-noise ratio and the effect on tracing emission sources. Details could be found in the Responses to Referee 1 and the section 3.4.1 in revised manuscript.

Reviewer 2 questions the time-representativity of your data set. He also makes an interesting comment regarding the radiative impact of BC in snow.

R: Generally, the sampling sites were selected 50 km away from cities and at least 1 km upwind of the approach road or railway to minimize the effect of pollution from local sources across northwestern China. But we also agree with the reviewer that understanding spatial and temporal differences of ILAPs in snow is still challenges. Therefore, the comparison of the seasonal and interannual variability of the ILAPs in snow was investigated, and the result shows that the differences of ILAPs in snow are relatively small in the Arctic and northeastern China (Doherty et al., 2010; Wang et al.,

2013, 2017). We note that further snow field campaigns were still performed worldwide to limit the uncertainties of ILAPs in snow due to the spatial and temporal differences across northern China, the Himalayas, North America, Greenland and the Arctic since 1980s (Cong et al., 2015; Dang and Hegg, 2014; Doherty et al., 2010, 2014; Hegg et al., 2009, 2010; Huang et al., 2011; Xu et al., 2009, 2012; Zhao et al., 2014; Warren and Wiscombe, 1980, 1985). For instance, a similar paper on the mixing ratios of ILAPs in Arctic snow has been widely used for validating modeled snow BC mixing ratios (Doherty et al., 2010). Therefore, we indicated that the datasets in this study can contribute to advancing remote sensing techniques and reducing the uncertainties of the model simulations to enhance our further understanding of the climate impacts of ILAPs in snow and ice.

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**Properties of black carbon and other insoluble  
light-absorbing particles in seasonal snow of northwest  
China**

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**Abstract.** A large field campaign was conducted ~~in northwestern China from January to February 2012~~ and 284 snow samples were collected at 38 sites in Xinjiang Province and 6 sites in Qinghai Province across northwestern China from January to February 2012. A spectrophotometer combined with chemical analysis was used to measure the insoluble light-absorbing particles (ILAPs) and chemical ~~components~~ species in ~~the seasonal~~ snow. ~~The results indicate that and two hundred eighty four snow samples were collected at 38 sites in Xinjiang Province and 6 sites in Qinghai Province.~~ The cleanest snow was found in northeastern Xinjiang along the border of China, and it presented an estimated black carbon ~~(BC)~~ ( $C_{BC}^{est}$ ) of approximately 5 ng g<sup>-1</sup>. The dirtiest snow ~~was found near industrial cities, and it presented a  $C_{BC}^{est}$  of approximately 450 ng g<sup>-1</sup>~~ near industrial cities in Xinjiang. Overall, the  $C_{BC}^{est}$  of most of the snow samples collected in this campaign was in the ranges of 10-150 ng g<sup>-1</sup>. Vertical variations in the snowpack ILAPs indicated a probable shift in emission sources with the progression of winter. An analysis of the fractional contributions to absorption implied that organic carbon (OC) dominated the 450-nm absorption in Qinghai, ~~whereas while~~ the contributions from BC and OC were comparable in Xinjiang. ~~Finally, a~~ Positive Matrix Factorization (PMF) model was run to explore the sources of particulate light absorption, and the results indicated an optimal ~~three~~ 3-factor/source solution that included industrial pollution, biomass burning, and soil dust. ~~In addition, the chemical components were evaluated to examine the mass contributions. In Qinghai, biomass burning was the dominant absorption factor despite the high mass contribution from soil dust. In Xinjiang, the~~

~~primary absorption source was industrial pollution at sites near cities and biomass burning at most sites in other regions. A negative correlation was observed between the BC mixing ratio and altitude in Xinjiang. An analysis based on the PMF three3-factor solution showed that this relationship likely resulted from gradient variations in the contributions of industrial pollution sources.~~

## 1 Introduction

The deposition of insoluble light-absorbing particles (ILAPs), primarily black carbon (BC), organic carbon (OC), and dust, on snow can reduce snow albedo (Warren and Wiscombe, 1980; Chylek et al., 1983; Brandt et al., 2011; Hadley and Kirchstetter, 2012), which can significantly affect regional and global climate (Jacobson, 2002, 2004; Hansen and Nazarenko, 2004; Flanner et al., 2007, 2009; McConnell et al., 2007; Ramanathan and Carmichael, 2008; Bond et al., 2013). [Warren and Wiscombe \(1980\)](#) indicated that a mixing ratio of  $10 \text{ ng g}^{-1}$  of BC in snow with snow grain size of  $1000 \text{ }\mu\text{m}$  may reduce the snow albedo at 400 nm by approximately 1%. ~~Warren and Wiscombe (1980) suggested that a mixing ratio of  $10 \text{ ng g}^{-1}$  of BC in snow may reduce the snow albedo by approximately 1%.~~ A modeling study indicated that soot could reduce snow and sea ice albedo by 0.4% from the global average and by 1% in



the Northern Hemisphere (Jacobson, 2004). Previous studies found that the “efficacy” of BC/snow forcing in the Arctic is more than three times greater than that of forcing by CO<sub>2</sub> (Hansen and Nazarenko, 2004; Flanner et al., 2007). However, the regional and global radiative forcing affected by the ILAPs in snow/ice is highly uncertain still a challenge~~However, radiative forcing is highly uncertain.~~ For instanceexample, Hansen and Nazarenko (2004) found that the effect of soot on snow and ice albedo yielded a climate forcing of +0.3 W m<sup>-2</sup> in the Northern Hemisphere. Recently, the IPCC’s AR5 (2013) reported that the radiative forcing from BC in snow and ice is 0.04 W m<sup>-2</sup> of the global mean, although it presents a low confidence level. Bond et al. (2013) indicated that the best estimate of climate forcing from BC deposition on snow and sea ice in the industrial era is +0.13 W m<sup>-2</sup> with 90% uncertainty bounds of +0.04 to +0.33 W m<sup>-2</sup>. The all-source present-day climate forcing including preindustrial emissions is somewhat higher at +0.16 W m<sup>-2</sup>.~~(Flanner et al., 2007? or 2009?, should cite literature here).~~ However, recent studies indicated that mMany factors complicate the evaluation of climate forcing by BC in snow (Hansen and Nazarenko, 2004; Bond et al., 2013). Hence, abundant comprehensive field campaigns are required to collect snow samples and measure ILAPs in snow to limit this uncertainty.

Recently, a number of field campaigns have been conducted to measure the BC in snow and ice in the Arctic (Clarke and Noone, 1985; Chylek et al., 1987, 1995; Cachier and Pertuisot, 1994; Grenfell et al., 2002; Hagler et al., 2007a, 2007b; McConnell et al., 2007; Forsstrom et al., 2009; Doherty et al., 2010, 2013). Overall, the BC mixing ratios in the snow of the Arctic were 3-30 ng g<sup>-1</sup>. The cleanest snow

was found on the Greenland Ice Sheet, whereas the dirtiest snow was found in East Russia (Doherty et al., 2010). Modeling studies have also evaluated the effect of BC in snow on warming in the Arctic (Hansen and Nazarenko, 2004; Jacobson, 2004; Flanner et al., 2007, 2009; Koch et al., 2009; Shindell and Faluvegi, 2009). Hansen and Nazarenko (2004) suggested that the mean effect of soot on the spectrally integrated albedos in the Arctic is 1.5%. Flanner et al. (2007) noted that the simulated annual Arctic warming is 1.61°C and 0.50°C for 1998 and 2001 central experiments when BC is included in snow compared with control simulations without BC. However, limited field campaigns and modeling studies are available in North America (Chylek et al., 1987; Qian et al., 2009; Hadley et al., 2010; Dang and Hegg, 2014; Doherty et al., 2014), Europe (Eleftheriadis et al., 2009; Thevenon et al., 2009; Painter et al., 2013; Gabbi et al., 2015; Peltoniemi et al., 2015), the Tibetan Plateau (Xu et al., 2006, 2009, 2012; Kang et al., 2007; Ming et al., 2008, 2009; Qiu, 2008; Cong et al., 2013; Wang et al., 2014; Li et al., 2016), and North China (Huang et al., 2011; Ye et al., 2012; Wang et al., 2013; Zhao et al., 2014), where snow is closer to the sources of ILAPs and more exposed to sunlight; therefore, the effect of ILAPs on snow may be more significant. Hence, we conducted a large field campaign ~~across northwestern China from January to February 2012~~ to measure the ILAPs in seasonal snow across northwestern China from January to February 2012.

In addition to BC, which presented the most absorptive impurity per unit mass, OC and dust can also significantly contribute to particulate light absorption in snow. OC in snow may be related to either combustion products that are deposited onto snow or

soil that is mixed into snow. Xu et al. (2006) first quantified the OC content on the Tibetan Plateau and determined the effect of OC on surface snow melting. Wang et al. (2013) suggested that OC dominates the particulate light absorption across the grasslands of Inner Mongolia in North China. 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)~~Light absorption by dust is usually related to iron oxides. Although its ability to reduce snow albedo is less than that of BC by approximately a factor of 50 (Warren, 1984), dust is the dominant absorber in snow locations.~~ For example, the increased radiative forcing by dust in snow has affected the timing and magnitude of runoff from the Upper Colorado River Basin (Painter et al., 2007, 2010).

Understanding the sources of ILAPs in snow is necessary for examining the climatic effects of ILAPs in snow. Certain scientists have focused on exploring the potential sources of BC in snow (Flanner et al., 2007; Shindell et al., 2008; Forsstrom et al., 2009); however, these studies primarily relied on numerical transport modeling based on limited data sets from emission inventories or calculated back trajectories, and they showed a limited ability to attribute the sources~~quantify the source attribution~~ of BC. Recently, a standard receptor model has been successfully used to resolve the sources of snow-BC ILAPs in snow. For example, Hegg et al. (2009, 2010) utilized measured

ILAPs and chemical components as inputs to run the model. The results showed four sources (crop and grass burning, boreal biomass burning, pollution, and marine) of light-absorbing particles in the Arctic snow. Biomass burning sources dominated during spring, although pollution played a more significant role during fall and winter and in summer in Greenland. Zhang et al. (2013a) evaluated the source attribution of ILAPs in the snow in northeastern China and found three sources for the ILAPs in the snow pack: soil dust (53%), industrial/urban pollution (27%), and biomass and biofuel burning (20%). These authors concluded that soil dust was the dominant source in the northeastern area of China, which is consistent with assessments that were based on back trajectory cluster analyses. A similar study by Doherty et al. (2014) focused on the sources of snow BC in North America and found that both biomass and fossil fuel combustion were the main sources of snow BC in the Pacific Northwest, Intermountain West and Canada, whereas soil dust played a predominant role in particulate light absorption in the northern U.S. Plains. Obviously, analyses based on the receptor model are timely and useful for identifying the source attribution of snow BC; thus, this model can be used to assess the potential sources of ILAPs in the snow in northwestern China because of the variety of emission sources (Ye et al., 2012).

In this study, we analyzed the spatial and vertical distributions of ILAPs in the seasonal snow ~~in~~ across northwestern China ~~to~~; ~~investigate~~ the contributions from BC, OC and Fe to particulate light absorption. The chemical components and ILAPs were also used to ~~and~~ estimated the potential emission sources and source attributions of ~~snow-BC~~ ILAPs in seasonal snow. ~~Finally, we evaluated the chemical components~~

~~to examine the potential emission sources.~~

## 2 Methods

### 2.1 Snow collection

Seasonal snow was collected at 38 sites in Xinjiang and 6 sites in Qinghai in China from January to February 2012, with 284 snow samples obtained across the entire expedition. Figure 1 shows the locations of the sampling sites, which were numbered in chronological order and followed the field campaign by Wang et al. (2013). Fresh snow was gathered from 13 sites where snow was falling at the time of sampling. ~~As shown in Figure 1 (check the format of “Figure 1 or figure 1, or Fig. 1” in TC),~~

Forty-two sites were separated into 5 regions according to their geographical distribution to investigate the spatial variations in snow-containing contaminants and their potential sources, with ~~the one rRegion 1 (Region 1) located~~ in Qinghai and ~~the other rRegions of (Regions 2-5) located~~ in Xinjiang ~~(Figure 1)~~.

Normally, snow ~~samples wereas~~ collected at vertical intervals of 5 cm from the top to the bottom at each site. If distinct layering was present, such as a melt layer or a top layer of newly fallen snow, that layer was ~~sampled collected~~ individually. In Qinghai, the snow was thin and patchy at sites 47-49; therefore, the samples were collected from drift snow. ~~Left and right snow samples were gathered within each layer in two adjacent vertical profiles to compare and average the sample pairs. 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).The snow density and temperature were also measured within each layer to~~

~~quantify the deposition flux of the BC or other ILAPs.~~ The sampling sites were selected 50 km far from cities and at least 1 km upwind of the approach road or railway to minimize the effect of pollution from local sources and achieve a representation of large areas.

5 The snow samples were filtered at four temporary laboratories to prevent the melting snow from influencing the ILAP<sub>s</sub> content. The snow samples were quickly melted in a microwave oven and then immediately filtered through a 0.4- $\mu$ m Nuclepore filter. The samples “before” and “after” filtration were collected and refrozen for subsequent chemical analyses, and the filters were used to measure the light absorption of  
10 ILAP<sub>s</sub>~~the filters were subjected to BC and OC analyses.~~ Additional details on the snow collection and filtration processes have been previously reported (Doherty et al. 2010, 2014; Wang et al., 2013).

## 2.2 Chemical speciation

The chemical analysis performed here followed that of Wang et al. (2015) and was  
15 similar to the procedures described by Zhang et al. (2013a) and Doherty et al. (2014). Details of these approaches have been previously reported fromby (check use “from”  
or “by” in TC) (Yesubabu et al., (2014). Briefly, the major ions ( $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ ,  $\text{Cl}^-$ ,  $\text{F}^-$ ,  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$  and  $\text{NH}_4^+$ ) were analyzed with an ion chromatograph (Dionex, Sunnyvale, CA, USA), and the trace elements (e.g., Fe, Al, Cu, Mn, Cr, and Ba) were  
20 measured by inductively coupled plasma mass spectrometry (ICP-MS). Pairs of unfiltered and filtered snow water samples were analyzed to evaluate the possible effect of filtering on the chemical constituents, and obvious differences were not

observed. Mineral dust (MD), Cl salt, biosmoke K ( $K_{\text{Biosmoke}}$ ) and trace element oxides (TEO) were determined to assess the mass contributions of the major components in the surface snow. The mineral dust content was calculated by a straightforward method, and the Al concentration in dust was estimated at 7% (Zhang et al., 2013b):

$$\text{MD}=\text{Al}/0.07 \quad (1)$$

Cl salt was estimated as follows in accordance with Pio et al. (2007) and Zhang et al. (2013b):

$$\begin{aligned} \text{Cl salt} &= \text{Na}_{\text{Ss}}^+ + \text{Cl}^- + \text{Mg}_{\text{Ss}}^{2+} + \text{Ca}_{\text{Ss}}^{2+} + \text{K}_{\text{Ss}}^+ + \text{SO}_{4\text{Ss}}^{2-} \\ &= \text{Na}_{\text{Ss}}^+ + \text{Cl}^- + 0.12\text{Na}_{\text{Ss}}^+ + 0.038\text{Na}_{\text{Ss}}^+ + 0.038\text{Na}_{\text{Ss}}^+ + 0.25\text{Na}_{\text{Ss}}^+ \end{aligned} \quad (2)$$

where  $\text{Na}_{\text{Ss}}^+$  is sea salt  $\text{Na}^+$ , 0.12, 0.038, 0.038, and 0.25 are the mass ratios in seawater of magnesium to sodium, calcium to sodium, potassium to sodium and sulfate to sodium, respectively.  $\text{Na}_{\text{Ss}}^+$  is calculated as follows (Hsu et al., 2009):

$$\text{Na}_{\text{Ss}} = \text{Na}_{\text{Total}} - \text{Al} * (\text{Na}/\text{Al})_{\text{Crust}} \quad (3)$$

where  $(\text{Na}/\text{Al})_{\text{Crust}}$  is the Na/Al ratio of representative dust materials (Wedepohl, 1995). Following Hsu et al. (2009), we estimated all three fractions (dust, sea salt, and biosmoke fractions) of K in snow, and  $K_{\text{Biosmoke}}$  was determined as follows:

$$K_{\text{Biosmoke}} = K_{\text{Total}} - K_{\text{Dust}} - K_{\text{Ss}} \quad (4)$$

$$K_{\text{Dust}} = \text{Al} * (\text{K}/\text{Al})_{\text{Crust}} \quad (5)$$

$$K_{\text{Ss}} = \text{Na}_{\text{Ss}} * 0.038 \quad (6)$$

where  $(\text{K}/\text{Al})_{\text{Crust}}$  is 0.37 and represents the K/Al ratio in the dust materials (Wedepohl, 1995) and  $\text{Na}_{\text{Ss}}$  is estimated by Equation (3). Following Zhang et al.

(2013b), we calculated the contribution of TEO using the following equation:

$$\text{TEO} = 1.3 * \sum_i (\alpha_i * \text{TE}_i) \quad (7)$$

where TE represents the trace elements (Ti, V, Cr, Mn, Co, Ni, Cu, Zn, As, Se, Sr, Mo, Ag, Cd, Sn, Ba, Hg, Tl, Pb, Th, U, and Be) determined via ICP-MS and  $\alpha$  is the weight coefficient of each trace element.  $\alpha$  is a function of the enrichment factor (EF):

$$\alpha = 0, \quad \text{EF} \leq 1;$$

$$\alpha = 0.5, \quad 1 < \text{EF} < 5; \quad (8)$$

$$\alpha = 1, \quad \text{EF} \geq 5.$$

where the EF of a given element (E) is calculated by the equation  $\text{EF} = (\text{E}/\text{Al})_{\text{Snow}} / (\text{E}/\text{Al})_{\text{Crust}}$  (Hsu et al., 2010).  $(\text{E}/\text{Al})_{\text{Snow}}$  and  $(\text{E}/\text{Al})_{\text{Crust}}$  are the ratios of the elements to the Al concentration in the snow sample and crust (Wedepohl, 1995), respectively. A multiplicative factor of 1.3 was used to convert the element abundance to the oxide abundance, which is similar to the method of Landis et al. (2001).

### 2.3 Spectrophotometric analysis

The filters were analyzed for the ILAPs ~~(check “ILAPs” in the whole manuscript)~~ content in the snow using a modified integrating-sandwich spectrophotometer (ISSW), which was described by Grenfell et al. (2011) and ~~used-performed~~ by Doherty et al. (2010, 2013, 2014, 2015), Dang and Hegg (2014) and Wang et al. (2013, 2015). This ISSW spectrophotometer measures the light attenuation spectrum from 400 to 700 nm where the signal-to-noise ratio is optimized. The total light attenuation spectrum is



extended over the full spectral range by linear extrapolation from 400 to 300 nm and from 700 to 750 nm (Grenfell et al., 2011). Light attenuation is nominally only sensitive to ILAPs on the filter because of the diffuse radiation field and the sandwich structure of two integrated spheres in the ISSW (Doherty et al., 2014). By considering all of the light absorption that occurred from 650-700 nm, the maximum possible BC ( $C_{BC}^{max}$ ) mixing ratio was calculated by calibrating the results against a set of fullerene (Alfa Aesar, Inc., Ward Hill, MA, USA) standard filters with a mass absorbing coefficient (MAC) of  $6.3 \text{ m}^2 \text{ g}^{-1}$  at 550 nm. In general, BC, OC and dust (Fe) dominated the light absorption by ILAPs in the snow samples. The equivalent BC ( $C_{BC}^{equiv}$ ), estimated BC ( $C_{BC}^{est}$ ), absorption Ångström exponent of all ILAPs ( $\text{\AA}_{tot}$ ), and light absorption fraction by non-BC ILAPs ( $f_{nonBC}^{est}$ ) were calculated by using the wavelength dependence of the measured spectral light absorption and by assuming that the MACs of the BC, OC and Fe were 6.3, 0.3, and  $0.9 \text{ m}^2 \text{ g}^{-1}$  at 550 nm, respectively, and that the absorption Ångström exponents ( $\text{\AA}$  or AAE) were 1.1, 6, and 3, respectively. The definitionsdescription of these parametersvariables can be found in Table S1. The details of this analysis were interpreted according to Grenfell et al. (2011). The OC mixing ratio was also determined according to Equation (2) in Wang et al. (2013), and the Fe concentration was determined according to the ICP-MS measurements.

Many studies (e.g., Jacobson, 2001; Hadley and Kirchstetter, 2012; Bond et al., 2013) have indicated that the MAC of BC is somewhat higher than the value used here, and Bond and Bergstrom (2006) recommended a value of  $7.5 \pm 1.2 \text{ m}^2 \text{ g}^{-1}$  at 550 nm.

However, we applied a value of  $6.3 \text{ m}^2 \text{ g}^{-1}$  to provide a comparison with previous studies (Hegg et al., 2009, 2010; Doherty et al., 2010; Wang et al., 2013). If the MAC of BC is actually close to  $7.5 \text{ m}^2 \text{ g}^{-1}$ , then our measured mass mixing ratio will be too high by a factor of 1.19. If the radiation models are run with the BC mixing ratio reported in this study, then the MAC of BC must be  $6.3 \text{ m}^2 \text{ g}^{-1}$ ; otherwise, the BC mixing ratio should be scaled appropriately.

#### 2.4 PMF model

The Positive Matrix Factorization (PMF) model that was used here (US EPA PMF 5.0) is a receptor model that provides source attribution by identifying and quantifying source profiles and contributions prior, which is based on mathematical approaches~~can quantify contributions from sources to samples based on the composition or fingerprints of the sources~~, and it has been widely applied (e.g., Amato et al., 2009; Amato and Hopke, 2012). The speciation or composition is determined by using analytical methods appropriate for the media, and key component species~~or combinations of components species~~ are required to distinguish the effects. The PMF model is a multivariate factor analysis tool that decomposes a matrix of speciated sample data into two matrices: factor contributions and factor profiles. These factor profiles must be interpreted by the analyst to identify the source types that may have contributed to the sample by using available ancillary information, such as the measured source profile information and emission or discharge inventories. The characteristic factor profiles are completely dependent on the mathematical approaches of the PMF model; 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.

~~therefore, the number of factors is not known a priori and must be selected individually in terms of the analyst's understanding of the sources that affect the samples as well as the number of samples and species' characteristics.~~

~~The PMF model uses two data sets as inputs to weigh individual points. One is the set of the concentrations of the input species, including the chemically analyzed constituents, along with  $C_{BC}^{max}$ , and the other is the associated uncertainty data set.~~

~~Uncertainty estimates of the chemical concentrations are based on analyses of replicate standards with uncertainties that are calculated as twice the standard error of the mean for each analyzed species. The uncertainties of  $C_{BC}^{max}$  were calculated as per Doherty et al. (2010).  $C_{BC}^{max}$  was likely biased by errors in the assumed MAC of our fullerene standards, which was applied to convert the measured absorption to a maximum BC mass. However, the relative biases of  $C_{BC}^{max}$  were uniform across all of the data sets, and the results of the PMF analysis were dependent on the relative variance of the given species against the absolute concentration; therefore, the effects can be appropriately ignored (Doherty et al., 2014).~~

Normally, the PMF model is applied to analyze a time series of componentsspecies

concentrations at a single observation site to estimate temporal variations. In this study, we used the PMF model to analyze the spatial variations in source contributions.

Although atypical, previous studies have effectively employed this model and

confirmed its reliability in terms of factor analyses of spatial distributions (Paatero et al., 2003; Chen et al., 2007; Hegg et al., 2009, 2010; Zhang et al., 2013a; Doherty et al., 2014).

### 3 Results

5 Table 2 summarizes the 2012 northwestern China field campaign and lists the values for the  $C_{BC}^{equiv}$ ,  $C_{BC}^{max}$ ,  $C_{BC}^{est}$ ,  $A_{tot}$ , and  $f_{nonBC}^{est}$  at every sampling layer for each site. The cleanest snow was found in the far north of Xinjiang Province (sites 70, 75, 77 and 78) and the high-altitude sites in the Tianshan Mountains (sites 79 and 82) because these sites were located far from the emission sources; moreover, the melting-amplified  
10 effect on the BC mixing ratios was negligible because the field campaign was conducted in January and February when the snow had not yet melted. Although the  $C_{BC}^{est}$  result was low and reached values of 1 ng g<sup>-1</sup> at site 77, the  $C_{BC}^{est}$  estimates were considered too uncertain when the corresponding  $f_{nonBC}^{est}$  was > 85% according to Doherty et al. (2014); therefore, we did not consider these values. Thus, the lowest  
15  $C_{BC}^{est}$  values were approximately 5 ng g<sup>-1</sup>, which were smaller than the minimum BC mixing ratio of approximately 40 ng g<sup>-1</sup> measured in North China via the same spectrophotometric analysis (ISSW) (Huang et al., 2011; Wang et al., 2013) and comparable to the values of approximately 3 ng g<sup>-1</sup> from the Greenland Ice Sheet (Doherty et al., 2010). The highest  $C_{BC}^{est}$  was found at sites 53, 60, 67, 83 and 84. At  
20 site 83, the  $C_{BC}^{est}$  reached 619 ng g<sup>-1</sup> at the bottom layer; however, the underlying soil may have been responsible for this high value. Therefore, we note that this value should cannot be used to represent the regional background level of BC. [After](#)

~~excluding the bottom value of  $C_{BC}^{est}$  at site 83~~After ~~excluding site 83~~, the highest  $C_{BC}^{est}$  value was approximately  $450 \text{ ng g}^{-1}$ , which was much lower than the values of  $>1000 \text{ ng g}^{-1}$  in snow in the industrial area of northeastern China (Wang et al., 2013). Overall, the  $C_{BC}^{est}$  of most of the snow samples ranged from  $10\text{-}150 \text{ ng g}^{-1}$ , which were similar to the ~~visible~~-values by visual estimates reported by Ye et al. (2012), and BC measurements of  $4\text{-}120 \text{ ng g}^{-1}$  recorded from glaciers in Tibet and Xinjiang by ~~the~~ previous field campaigns that used a controlled combustion method (Xu et al., 2006, 2009; Ming et al., 2008, 2009).

### 3.1 Results by region

As discussed above, the sample sites were separated into five regions. Table ~~32~~ lists the regional averages and standard deviations of  $C_{BC}^{equiv}$ ,  $C_{BC}^{max}$ ,  $C_{BC}^{est}$ ,  $\dot{A}_{tot}$ , and  $f_{nonBC}^{est}$  for the surface and subsurface layers. The spatial distributions of  $C_{BC}^{est}$  and  $\dot{A}_{tot}$  for the surface snow samples are shown in Figure 2a and 2b. In addition, Figure 2c shows the equivalent BC value used to explain the 300-750-nm absorption by all non-BC ILAPs, which was calculated as  $C_{BC}^{equiv} * f_{nonBC}^{est}$ . Thus, Figures 2a and 2c can be compared to determine the relative contributions from BC and non-BC ILAPs to the snow albedo reduction at each site. However, the BC mass deposited onto snow over a specified period is more useful when comparing models than the surface values because the average BC over many snowfall events across a typical month or season presents a more representative contribution to the background levels throughout the entire accumulation period (Doherty et al., 2014). Thus, ~~in Table 3, we list~~ the integrated snow water equivalent (SWE) and the total BC mass for a  $1\text{-cm}^2$  column of

snow (integrated BC) were given in table 4. We also estimated the average BC mixing ratios in the snow column ( $\overline{C_{BC}^{est}}$ ), which were calculated as the integrated BC divided by the SWE (Figure 2d and Table 43). Indeed, the  $\overline{C_{BC}^{est}}$  value was more spatially uniform than the  $C_{BC}^{est}$  value.

5 In Region 1 (sites 47-52), which is located in the eastern Tibetan Plateau in Qinghai Province, the snow was thin and patchy and presented a sample snow depth of 2.5-10 cm. During windy periods, local soil can be lofted and deposited onto snow surface, and this deposition has been confirmed by previous reports (Ye et al., 2012), which ~~observed~~ shown as yellowed filters because of the heavy loading of soil dust.

10 Although the  $C_{BC}^{est}$  was intermediate and presented typical values of 30-150 ng g<sup>-1</sup>, both the  $C_{BC}^{equiv}$  and  $C_{BC}^{equiv} * f_{nonBC}^{est}$  values were highest in the surface (307±119, 213±89 ng g<sup>-1</sup>) and subsurface (332±201, 214±116 ng g<sup>-1</sup>) snow among all five regions (Table 32 and Figure 2) when considering the primary proportion of non-BC ILAPs in the mass of the ILAPs. In addition, the  $\hat{A}_{tot}$  values were 3.6-4.6 and higher  
15 than those in any other region, which indicated the predominant contribution from non-BC ILAPs to light absorption.

Region 2 (sites 53-59, 61, and 79) is located on the Tianshan Mountains, and except for site 58, all of the sites were located far from cities. Site 58, which is situated in the foothills near Yili pPrefecture, likely experiences a greater influence of human  
20 activities. All of the regional statistics were intermediate. The  $C_{BC}^{est}$  values were generally in the range of 20-100 ng g<sup>-1</sup>. At site 54, the surface  $C_{BC}^{est}$  was low and reached 7 ng g<sup>-1</sup>. Higher altitudes (> 3000 m) and freshly fallen snow at the surface

may have been responsible for this lower value. The higher mixing ratios at site 53 for both surface and subsurface snow may have been caused by local soil dust or biomass burning as reported by Ye et al. (2012). The  $\hat{A}_{\text{tot}}$  values varied remarkably from 2.2 to 4.5.

5 Region 3 (sites 60, 62-63, and 80-84) is located to the north of the Tianshan Mountains, and industrial cities are located close to this region, and the population density is much higher than that in any of the other regions; therefore, human activities may have dominated the contributions to ILAPs in the snow. However, the  $C_{\text{BC}}^{\text{est}}$  values, which primarily ranged from 10-100 ng g<sup>-1</sup>, were comparable to those in  
10 other regions in Xinjiang, which were inconsistent with the findings in North China, where the  $C_{\text{BC}}^{\text{est}}$  in the industrial northeastern area was higher by a factor of 10 compared with the values to the remote northeast on the border of Siberia (Wang et al., 2013). The  $f_{\text{nonBC}}^{\text{est}}$  for the surface and subsurface samples was considerable different and presented values of 58±15% and 72±12%, respectively, which reflects the  
15 temporal changes in the contributions from BC and non-BC particles to light absorption over time. The  $\hat{A}_{\text{tot}}$  values were typically 2-4, which were comparable to those in Region 2, despite the snow samples may have been more affected by human activities.

Region 4 (sites 64-70) is located in northwestern Xinjiang along the border of China.

20 The  $C_{\text{BC}}^{\text{est}}$  generally ranged from 20-150 ng g<sup>-1</sup>. The regional average  $\hat{A}_{\text{tot}}$  was approximately 3. The  $f_{\text{nonBC}}^{\text{est}}$  in this region was the lowest and presented an average value near 50%; therefore, the BC and non-BC particles presented almost identical

contributions to light absorption, which was inconsistent with the other regions, where non-BCs played a dominant role.

The cleanest snow of this campaign was found in Region 5 (sites 72-78) in northeastern Xinjiang. Most of the snow samples had low  $C_{BC}^{est}$  values of 10-50 ng g<sup>-1</sup>, which were much smaller than the values of 50-150 ng g<sup>-1</sup> in the cleanest snow in northeastern China. The  $\dot{A}_{tot}$  value generally ranged from 2.5 to 3.5 and presented a regional average of approximately 3, which was consistent with other regions in Xinjiang. The  $f_{nonBC}^{est}$  varied obviously from < 50% to > 90%. This wide range indicates the spatial variance in the dominant emission sources of particulate light absorption in this region.

### 3.2 Vertical variations in snowpack light-absorbing particulates

The vertical profiles of the  $C_{BC}^{max}$ ,  $C_{BC}^{est}$ , and  $\dot{A}_{tot}$  at each sample site are shown in Figure 3. At sites 49-52 in Qinghai (Region 1), an obvious dust layer was present in each site, and analyses of the ILAPs content by ISSW presented high uncertainties.

Therefore, we did not report the values at these layers. In addition, the sampling at sites 47-49 was conducted in drift snow as discussed above. Thus, the vertical profiles from these sites in Qinghai did not accurately represent the temporal variations in the deposition of snow, [although apparent vertical differences could be observed at could](#)

[sites](#)~~although apparent vertical differences were observed at could sites~~, such as site 47. In Regions 3, 4 and 5 in Xinjiang, the  $C_{BC}^{est}$  values were much larger in the surface snow (127±158, 126±124, and 74±56 ng g<sup>-1</sup>, respectively) than in the subsurface snow (75±120, 82±56, and 37±31 ng g<sup>-1</sup>, respectively), with the ratio of the  $C_{BC}^{est}$  from the



top layer to the average  $C_{BC}^{est}$  from all the subsurface layers presenting values of 1.7, 1.5, and 2, respectively, which indicates an increase in aerosol deposition later in winter. However, the  $C_{BC}^{est}$  values in the surface ( $81 \pm 102 \text{ ng g}^{-1}$ ) and subsurface layers ( $89 \pm 69 \text{ ng g}^{-1}$ ) in Region 2 were comparable. These differences in deposition could have been caused by changes in the dry deposition and/or the mixing ratio of the ILAPs to the precipitation water content in snowfall. Doherty et al. (2013) studied the observed vertical redistribution of BC and other ILAPs in melting snow and noted that melt amplification generally appears within the top few centimeters of the snowpack, and it increases the BC mixing ratios of the surface snow. However, the high surface BC values reported here could not have originated from this mechanism because the temperature at these sites was not high enough to melt the snow. The  $\dot{A}_{tot}$  and  $f_{nonBC}^{est}$ , the values increased by 0.41 and 10% from the subsurface to the surface snow in Region 2, respectively, but decreased by 0.40 and 14% in Region 3, respectively. These variations may have been partly caused by a shift in emission sources as winter progressed, with a greater contribution to the ILAPs in snow from biomass burning in Region 2 and fossil fuel burning in Region 3 during this season.

### **3.3 Contributions to particulate light absorption by BC, OC and Fe**

Figure 4a shows the fractional contributions of BC, OC and Fe to light absorption at 450 nm for the surface snow samples at each site. In Qinghai (Region 1), OC dominated the absorption, and the fractional contribution reached approximately 70%. However, dust accounted for the main portion of the particulate mass, which was confirmed by the yellow color of the filters, and the fractional contribution from Fe to

absorption (only approximately 5%) was not significant. In Xinjiang, the fractional contributions from BC, OC and Fe were approximately 45%, 50%, and 5%, respectively, and their patterns did not vary greatly by region. At sites 58, 75, and 77, the fraction of light absorption from Fe exceeded 20%. This value was much higher than the values at the other sites, which may be explained by two possible reasons: (1) less BC and OC were contributed by biomass and fossil fuel burning and more Fe was contributed by dust, and (2) Fe was contributed by industrial emissions as well as dust. Wang et al. (2013) analyzed the snow particulate light absorption by ILAPs in the snow samples ~~in~~ across northeastern China and noted that OC could be composed of soil organics, combustion aerosols, and/or other biological organics. BC and OC could be produced via combustion sources, such as fossil fuel burning, open burning and biofuel burning. Thus, we could not precisely separate the contributions from different sources by analyzing the ILAPs. Hence, a more feasible method is required to evaluate and quantify the contributions from emission sources to particulate light absorption (see Section 3.4).

The relative contributions of these three ~~components~~ species are related to the values of  $\hat{A}_{\text{tot}}$  and  $\hat{A}_{\text{nonBC}}$ , which are shown in Figure 4b. Overall, the  $\hat{A}_{\text{nonBC}}$  values were almost in a narrow range of 5-6, which indicated that OC was the major component of non-BC ILAPs because the  $\hat{A}$  values of OC and Fe were 6 and 3, respectively. Three exceptions were sites 58, 75, and 77, which had  $\hat{A}_{\text{nonBC}}$  of  $< 5$ , and higher fractional contributions from Fe to absorption likely resulted in these lower values. Compared with  $\hat{A}_{\text{nonBC}}$ , the  $\hat{A}_{\text{tot}}$  values varied greatly from 1.84 to 4.66, and the

large variety of BC mixing ratios was mainly responsible for the large changes in  $\dot{A}_{\text{tot}}$ . In terms of the spatial distribution, the  $\dot{A}_{\text{tot}}$  value was close to the  $\dot{A}_{\text{nonBC}}$  value in Qinghai (Region 1), although the differences between  $\dot{A}_{\text{tot}}$  and  $\dot{A}_{\text{nonBC}}$  were highly variable in Xinjiang.

5 As discussed in Section 2.3, we attributed the snow particulate absorption to BC, OC, and Fe based on the absorption optical depth measured by ISSW [spectrophotometer](#) along with the chemical analyses of Fe and assumptions for the  $\dot{A}$  values and mass absorbing coefficients of BC, OC, and Fe. Furthermore,  $\dot{A}_{\text{nonBC}}$  was determined by the light absorption of OC and Fe. However, the assumptions presented large  
10 uncertainties, which may have introduced considerable errors in the values of BC, OC, and  $\dot{A}_{\text{nonBC}}$ . Doherty et al. (2010) analyzed the errors that originated from these assumptions and indicated a likelihood of uncertainty of < 50% based on liberal evaluations of these potential sources of errors. [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 \pm 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  
15 \(or OC\) 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](#)  
20

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<sup>-1</sup>, 1000 (150) ng g<sup>-1</sup> and 50 (50) ng g<sup>-1</sup> 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±0.3, 6±2 and 3±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.

## **3.4 PMF results**

### **3.4.1 PMF source profile PMF model optimization**

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 factors, 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  $C_{BC}^{max}$  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)

$C_{BC}^{max}$  represents the mass of BC, assuming all of the particulate light absorption (650-700 nm) is related to BC; (2)  $C_{BC}^{max}$  is only calculated based on the assumed MAC of BC; therefore, the errors of  $C_{BC}^{max}$  were the lowest among the studied variables. The chemical components were  $SO_4^{2-}$ ,  $NO_3^-$ ,  $Cl^-$ ,  $F^-$ ,  $Na^+$ ,  $K^+$ ,  $NH_4^+$ ,  $K_{Biosmoke}$ , 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  $C_{BC}^{max}$ ,  $Na^+$ ,  $K^+$  and Al, 50 % for  $SO_4^{2-}$ ,  $NO_3^-$  and  $K_{Biosmoke}$ , 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  $NO_3^-$ ,  $NH_4^+$ ,  $Cl^-$ ,  $F^-$ , 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  $R^2$  value for  $C_{BC}^{max}$  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.

### **3.4.2 PMF source profile**

The PMF 5.0 model was run using the chemical and spectrophotometric analysis data set of the surface snow samples. Following Doherty et al. (2014), we used  $C_{BC}^{max}$  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)  $C_{BC}^{max}$  represents the mass of BC, assuming all of the particulate light absorption (650–700 nm) is related to BC; and (2)  $C_{BC}^{max}$  is only calculated based on the assumed MAC of BC; therefore, the errors of  $C_{BC}^{max}$  were the lowest among the studied variables. Three to seven factors and 7 or more random seeds were always applied in the PMF model. Thus, the optimal number of factors/sources was 4 based on the robust and theoretical Q values (Hegg et al., 2009, 2010). However, 3 factors 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  $R^2$  value for  $C_{BC}^{max}$  with this 3-factor solution was considerably high (0.87). Rotational ambiguity was tested by varying the peak parameter, which also indicated stable results. Hence, the 3-factor solution was the best choice.

Figure 5 shows the source profile, including the calculated ~~measured~~ mass concentrations (lines) and the percent of components ~~species~~ apportioned to each factor (dots) for the three ~~3~~-factor solution. The first factor (top panel) was obviously characterized by high loadings of  $SO_4^{2-}$ ,  $NO_3^-$ ,  $Cl^-$ , and  $NH_4^+$ .  $SO_4^{2-}$  and  $NO_3^-$  are well-known markers for the burning of fossil fuel, such as coal and oil (e.g., Xie et al., 2008; Oh et al., 2011; Zhang et al., 2013b).  $Cl^-$  is usually regarded as an important component of sea salt but also a product of industrial emission (Kulkarni, 2009; Dall'Osto et al., 2013) and coal combustion (Hailin et al., 2008). Furthermore, the

molar ratio of  $\text{Cl}^-$  to  $\text{Na}^+$  was clearly larger than that of sea salt by a factor of  $> 2$ , which implied another source in addition to sea salt. Additionally,  $\text{NH}_4^+$  is a recognized marker of coal combustion. However, the ratio of  $\text{C}_{\text{BC}}^{\text{max}}$  to  $\text{SO}_4^{2-}$  was low (0.04) and close to the values in pollution sources as reported by Hegg et al. (2009).

5 Therefore, we considered the first factor an industrial pollution source. The second

factor (middle panel) presented substantial loadings of  $\text{Na}^+$ ,  $\text{K}^+$ , and  $\text{K}_{\text{Biosmoke}}$ .

However, chemical analyses are not available for certain organic matter, including levoglucosan, succinate, oxalate, and formate, which generally indicate biomass

burning.  $\text{K}^+$  and  $\text{K}_{\text{Biosmoke}}$  are markers for biomass burning emissions and were

10 highly loaded for this factor. In particular,  $\text{K}_{\text{Biosmoke}}$ , which was calculated as the

biosmoke fraction of K, was representative (Zhang et al., 2013a). Compared with the

first factor, the molar ratio of  $\text{Cl}^-$  to  $\text{Na}^+$  was smaller than 1, which indicates that

$\text{Na}^+$  was also a potential product of emission sources in addition to sea salt, such as biomass burning (Oh et al., 2011). However, the ratio of  $\text{C}_{\text{BC}}^{\text{max}}$  to  $\text{SO}_4^{2-}$  was

15 relatively high compared with that of the previously identified industrial pollution

sources and close to the ratio for a biomass burning source as reported by Doherty et

al. (2014). Therefore, we interpreted the second factor as a biomass burning source.

The third factor (bottom panel) accounted for over 50% Al, Cr, Fe, Cu, and Ba in the samples. Al and Fe are well-known crustal constituents, and they are usually used to

20 determine the mass of soil dust. The mass ratio of Fe to Al (0.36) was close to the

value in the continental crust (0.40) (Wedepohl, 1995). Furthermore, the enrichment

factors (EFs) of the trace elements, including Cr, Cu, and Ba, were  $< 5$  at many



sample sites, which indicates that these elements may have originated from a crustal source (Zhang et al., 2013b). Hence, we can interpret the third factor as a soil dust source.

#### **3.4.32 Source contributions to the sample sites**Source attribution in snow

5 Figure 6 shows the mean normalized contributions from each source to the individual receptor site. The industrial pollution source with a 31%  $C_{BC}^{max}$  dominated the contributions to the sites in Region 3. In particular, the mean normalized contributions were  $> 5$  at sites 60 and 84, which was consistent with the abundant loadings of  $SO_4^{2-}$  and  $NO_3^-$ . The results were not surprising because substantial industrial activity  
10 occurs in this region. However, the contributions from the biomass burning source with the highest  $C_{BC}^{max}$  (53%) were more geographically dispersed and relatively evenly distributed across Regions 1, 2, and 4. At sites 53 and 67, the contributions were significantly large, which were likely caused by the concentrations of  $K_{Biosmoke}$ , which were over  $500 \text{ ng g}^{-1}$  and much higher than those at the other sites. Indeed,  
15 biomass burning, such as biofuel combustion for heating, in winter and early spring in northwestern China is normally prevalent (Pu et al., 2015). Unsurprisingly, a soil dust source, which was characterized by the highest loadings of Al and Fe, was mainly associated with the sites in Qinghai (Region 1), although the contributions were obvious at certain sites in Xinjiang, especially the sites in Region 5, which was  
20 partially because these sites were located on hills with scarce plants, and wind may have blown local soil dust onto the snow.

#### **3.4.3 Source attribution of the particulate absorption**

The fractional contributions from the three sources to the 650-700-nm particulate absorption at each individual receptor site are shown in Figure 7. The average regional contributions are shown in Table 54. The most remarkable feature of the source attributions is the differences observed by region. Biomass burning was the primary source in Region 1 (in Qinghai) and in Regions 2 and 4 (in Xinjiang), and it presented average regional contributions of 59%, 60%, and 67%, respectively. Although high dust mass was present in the snow samples from Region 1, this source attribution was reasonable because the  $C_{BC}^{\max}$  from the biomass burning sources was much larger than that from the soil dust sources by a factor of  $> 3$  (Figure 6). In particular, biomass burning in Qinghai is prevalent, especially during winter (Yan et al., 2006). In Region 1, soil dust accounted for 29% of the particulate absorption, which was less than the contribution from the biomass burning sources but more significant compared with the contributions from soil dust in the other regions. In Regions 2 and 4, most of the sample sites were located on mountains and far from industrial areas; therefore, dominant absorption by biomass burning sources was not anomalous. The only exception was site 58, which was dominated by industrial pollution sources, and this result was likely because of its shorter distance from cities and lower elevation. In Region 3, all of the sample sites were located near cities and suffered from anthropogenic emissions; therefore, industrial pollution was the primary source and presented a contribution of 58%. In Region 5, the primary source differed between sites. Absorption was dominated by biomass burning sources at sites 73 and 78, by industrial pollution sources at sites 72, 74, and 76, and by soil dust sources at

sites 75 and 77. More complex topography and emission sources could partly explain these findings.

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.

5 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.  $K^+$  and  $K_{\text{Biosmoke}}$  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  $C_{\text{BC}}^{\text{max}}$  instead of ILAPs to estimate the

10 fractional contributions to the 650-700-nm particulate absorption~~The PMF results in Qinghai in this study were inconsistent with those by Zhang et al. (2013a), who indicated that soil dust was the dominant source of ILAPs. However, the discrepancy was mild because (1) the receptor sites were far from each other, (2) the chemical species inputs were different, and (3) the variables that characterized the particulate~~  
15 ~~light absorption in the PMF analysis were inconsistent.~~ Hence, additional sample sites

and more complete and advanced analyses of ILAPs and chemical components species inputs to the PMF model are necessary to obtain more representative source analysis results.

#### **3.4.44 Altitude gradients of BC mass**

20 Ye et al. (2012) performed a preliminary study on the same field campaign and found that the snow BC mixing ratios, which were based on visual estimates, were negatively correlated with the altitudes of the sample sites in Xinjiang. In our study,

the  $C_{BC}^{est}$  and  $C_{BC}^{max}$  values from the ISSW presented a similar trend (Figure 8a and b). Thus, altitude is an important influencing factor for BC mixing ratios and particulate absorption. Additionally, the  $C_{BC}^{max}$  value simulated by the PMF model decreased steadily with altitude, although this trend was not as obvious as that for the measured  $C_{BC}^{est}$  and  $C_{BC}^{max}$ . Therefore, we can explore the cause of altitude gradients for the BC mass based on the PMF ~~three~~-factor solution. Figure 9 shows the scaled contributions ~~contributions~~ of each source, which was obtained by multiplying the simulated contributions by the ratio of measured to calculated  $C_{BC}^{max}$ , as a function of altitude. We note that the snow samples with scaled contributions larger than 3 were removed due to heavy polluted snow samples (Ye et al., 2012)~~The sample snow from site 53 was dirty (Ye et al., 2012); therefore, we did not consider the results of that site in the trend analysis.~~ Clearly, the contributions from industrial pollution sources presented a decreasing trend with altitude and, was highly related to the altitude, while the biomass burning and soil dust only showed weak correlations~~whereas the contributions from biomass burning and dust soil sources did not show obvious gradient variations.~~ Thus, the altitude gradients of the contributions from industrial pollution likely caused the altitude gradients of the BC mass, ~~and the effects of biomass burning and soil dust were limited.~~

### 3.5 Mass contribution of the chemical components

In addition to performing a PMF analysis, the chemical components must be evaluated to examine the potential emission sources. As shown in Figure 10a, the median mass concentrations of chemicals in the different regions were  $6.0 \mu\text{g g}^{-1}$

(Region 1),  $5.0 \mu\text{g g}^{-1}$  (Region 2),  $7.0 \mu\text{g g}^{-1}$  (Region 3),  $5.0 \mu\text{g g}^{-1}$  (Region 4), and  $4.6 \mu\text{g g}^{-1}$  (Region 5). At sites 60 and 84, the mass concentrations were  $> 20 \mu\text{g g}^{-1}$ , which were primarily contributed by  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$ . Regionally, the chemical components were dominated by MD (30.5%) and OC (29.5%) in Region 1, MD (21.1%) and  $\text{SO}_4^{2-}$  (20.2%) in Region 2,  $\text{SO}_4^{2-}$  (21.5%) and  $\text{NO}_3^-$  (29.2%) in Region 3,  $\text{SO}_4^{2-}$  (17.9%) and  $\text{NO}_3^-$  (33.7%) in Region 4, and MD (29.5%) and  $\text{NO}_3^-$  (27.5%) in Region 5 (Figure 10b). These results indicated that soil dust sources greatly contributed to the chemical components in Region 1, whereas industrial pollution was the predominant source in Regions 3 and 4. In the other regions, the fractional contributions from different sources to the mass concentrations were comparable. The mass contribution was not proportional to the absorption contribution because of the different ILAPs loadings of emission sources. For example, although  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  are [major primary component species](#), the 650-700-nm particulate absorption in Region 4 was dominated by a biomass burning source. BC is regarded as an important light-absorbing particle, and it ranged from 0.2 to 4.8% in mass at all sites, with an average of 1.3%, which is smaller than that (2.4-5.1%) in urban areas in China (Huang et al., 2014).  $K_{\text{Biosmoke}}$  is a good indicator of biomass burning, and it ranged from 0.4 to 7.8% and presented the largest regional average fractional contribution (3.0%) in Region 2. Wang et al. (2016) reported comparable values of 1.3-5.1% in the snow in northeastern China.

### 3.6 Comparative analysis of chemical components

Bond et al. (2004) indicated that the OC:BC ratio of the emissions from fossil fuel

burning is lower than that from biomass or biofuel burning; therefore, we may qualitatively examine the primary emission sources based on this theory. In this work, the regional average ratios of OC to BC were 20.9, 6.12, 3.99, 6.71, and 7.28 (Figure 11a). The smallest value in Region 3 was similar to those observed in Beijing (Zhang et al., 2013b). The similar ratios in Regions 2, 4, and 5 were close to that of savanna and grassland regions as reported by Andreae and Merlet (2001). The results suggested the relative dominance of an industrial pollution source in Region 3 and a biomass burning source in other regions in Xinjiang. This pattern was similar to that of the source apportionment analysis by the PMF model. The largest value in Region 1 in Qinghai implied a primary contribution to OC from soil dust.

Sources of nitrate are considerably more varied than the sources of sulfate (Arimoto et al., 1996). For  $\text{NO}_3^-$ , the largest source is fossil fuel combustion. Biomass burning is regarded as another main source, which was determined according to the analysis of Logan. Additionally, microbial activity in soil is a potential source of nitrate. However,  $\text{SO}_4^{2-}$  is mainly a product of burning coal. Thus, we can compare the correlation between  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$  to explore the variety of emission sources. As shown in Figure 11b, the average regional ratio of  $\text{NO}_3^-$  to  $\text{SO}_4^{2-}$  in Region 1 was 0.49, which was lower than that in the other regions. The concentrations and correlation coefficients of  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$  were both the lowest in Region 1, which indicates limited emissions from an industrial pollution source. In Xinjiang, the ratios mostly ranged from 1 to 1.5, and the high correlation coefficient in Region 5 was associated with similar industrial pollution sources, whereas the low correlation coefficients in

Regions 2-4 may have been related to complicated industrial pollution sources.

A comparison between cations and anions is shown in Figure 11c. Generally, the correlation coefficients were all significant at the 5% level. A large anion charge deficit was observed in Region 1, which presented an average regional ratio of 2.93, which was likely caused by the absence of detected  $\text{CO}_3^{2-}$  and  $\text{HCO}_3^-$ . Carbonates (e.g.,  $\text{CaCO}_3$  and  $\text{MgCO}_3$ ) are often abundant in dust, which was observed in the Central Himalayan Glacier (Xu et al., 2013), and account for the largest contribution from soil dust to the mass concentration. However, the average ratios in the sample snow in Xinjiang were generally uniform and ranged from 0.7-1.2, which suggests an adequate charge balance. Overall, the concentrations of inorganic ions in the snow samples were lower than those in the rainwater in urban sites in China (Wang and Han, 2011) but larger than previous measurements in the Himalayas (Thompson et al., 2000; Xu et al., 2013).

#### **4 Discussion and Conclusions**

A large field campaign was conducted in northwestern China from January to February 2012, with 284 snow samples collected from 38 sites in Xinjiang Province and 6 sites in Qinghai Province. Based on a previous study in North China (Wang et al., 2013), we estimated the ILAP content and analyzed the chemical components. In addition, we used a PMF model to explore the fractional contributions from different sources to light absorption.

In Qinghai, the snow was thin and patchy; therefore, local soil could be lofted and deposited onto this snow. In this region, non-BC ILAPs predominantly contributed to

~~light absorption.~~ The cleanest snow of this campaign was found in the northeast of Xinjiang along the border of China, and the lowest  $C_{BC}^{est}$  values were approximately 5  $ng\ g^{-1}$ . The highest  $C_{BC}^{est}$  value was approximately 450  $ng\ g^{-1}$  at site 60, which was located in proximity to industrial cities. In Qinghai, OC dominated the 450-nm absorption, and the fractional contribution reached approximately 70%. Although dust accounted for the main portion of the particulate mass, the fractional contribution from Fe to absorption (approximately 5%) was not significant. In Xinjiang, the fractions of absorption from BC (45%) and OC (50%) were comparable, and the effect of Fe was limited. ~~Although the sites that were located to the north of the Tianshan Mountains were more affected by human activities, the average  $C_{BC}^{est}$  value was intermediate and comparable to the values in the other regions. Overall, the  $C_{BC}^{est}$  values of most of the snow samples ranged from 10 to 150  $ng\ g^{-1}$  in this campaign. In the Tianshan Mountains (Regions 2 and 3), the  $\dot{A}_{tot}$  and  $f_{nonBC}^{est}$  values presented vertical changes from the subsurface to surface snow, which indicates a probable shift in emission sources as winter progressed.~~

~~In Qinghai (Region 1), OC dominated the 450-nm absorption, and the fractional contribution reached approximately 70%. Although dust accounted for the main portion of the particulate mass, the fractional contribution from Fe to absorption (approximately 5%) was not significant. In Xinjiang, the fractions of absorption from BC (45%) and OC (50%) were comparable, and the effect of Fe was limited.~~

A [three](#)-factor solution (industrial pollution, biomass burning and soil dust) was used to explore the fractional contributions from the different sources within the particulate



absorption range from 650-700 nm based on a source apportionment analysis with the PMF model. In Qinghai, biomass burning was the primary source (59%), and soil dust accounted for 29% of the particulate absorption despite its high mass contribution. In Xinjiang, the source attributions varied by region. In Regions 2 and 4, most of the sample sites were located on mountains and far from industrial areas, and biomass burning sources were dominant and accounted for 60% and 67% of the contributions, respectively. In Region 3, absorption was dominated by an industrial pollution source (58%) because of the shorter distance from cities and lower elevations. In Region 5, the topography and emission sources were more complex, and the primary sources differed between sites. However, the BC mixing ratios showed a negative correlation with altitude in Xinjiang. ~~An analysis based on the PMF three-factor solution showed that this observation likely resulted from gradient variations in the contributions from industrial pollution sources., which likely resulted from gradient variations in the contributions of industrial pollution sources.~~

An evaluation indicated that the predominant source of chemical components was soil dust in Region 1, ~~while and~~ industrial pollution in Regions 3 and 4. The mass of BC, which is regarded as an important light-absorbing particle, ranged from 0.2 to 4.8% and presented an average of 1.3%. A comparison between OC and BC suggested the relative dominance of an industrial pollution source in Region 3 and a biomass burning source in other regions in Xinjiang. ~~The relationship between  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$  also showed limited emissions from an industrial pollution source in Qinghai but complicated sources in Xinjiang. Finally, a large anion charge deficit was observed in~~

~~Region 1, which explained the predominant contribution from soil dust to the mass concentration.~~

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## **5 Data availability**

All data sets and codes used to produce this study can be obtained by contacting Xin Wang (wxin@lzu.edu.cn). The elevation data used in this study is available: <http://rda.ucar.edu/datasets/ds759.3/#!access>.

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*Competing interests.* The authors declare that they have no conflict of interest.

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**Table 1.** Variables derived by using the ISSW spectrophotometer.

Symbols	Description of variables
$C_{BC}^{max}$	<u>Maximum BC is the mass of BC per mass of snow, if all particulate light absorption (650–700nm) is due to BC.</u>
$C_{BC}^{est}$	<u>Estimated BC is the estimated true mass of BC per mass of snow, derived by separating the spectrally resolved total light absorption.</u>
$C_{BC}^{equiv}$	<u>Equivalent BC is the amount of BC that would need to be present in the snow to account for the wavelength-integrated (300–750nm) total light absorption of down-welling solar radiation by all particulate constituents.</u>
$\mathring{A}_{tot}$	<u>Absorption Ångström exponent is calculated between 450 and 600nm, for all particulate deposited on the filter.</u>
$f_{nonBC}^{est}$	<u>Fraction of light absorption by non-BC ILAPs, is the absorption by non-BC particulate constituents, weighted by the down-welling solar flux, then spectrally integrated from 300 to 750nm.</u>

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**Table 24.** Statistics of the seasonal snow variables measured using an ISSW for each site.

Site	Layer	Latitude (N)	Longitude (E)	Average snow depth		Sample depth (cm)		$C_{BC}^{equiv}$ (ng g <sup>-1</sup> )	$C_{BC}^{max}$ (ng g <sup>-1</sup> )	$C_{BC}^{est}$ (ng g <sup>-1</sup> )	$f_{non-BC}^{est}$ (%)	$\hat{A}_{lot}$
				(cm)	depth	Top	Bottom					
47	1	35.54	99.49	2.5		0	5	480	242	148(—,235)	73(55,—)	4.58
	2					5	10	487	266	175(—,262)	64(46,—)	4.02
	3					10	15	1094	548	358(—,556)	67(49,—)	4.30
	4					15	20	314	200	156(29,217)	52(33,—)	3.55
48	1	34.85	98.13	4.5		0	1	334	196	152(65,222)	58(37,—)	3.87
	2					2	4	237	129	83(—,134)	65(43,—)	4.09
49	1	35.22	98.95	10		0	1	317	160	118(—,179)	66(47,—)	4.36
	2					2	7	—	—	—(—,—)	—(—,—)	—
50	3					11	16	315	156	102(—,164)	64(42,—)	4.38
	4					16	25	203	90	53(—,84)	74(59,—)	4.20
	1	34.80	99.05	—		0	2	165	66	28(—,54)	83(67,—)	4.57
	2					2	7	—	—	—(—,—)	—(—,—)	—
51a	3					7	12	—	—	—(—,—)	—(—,—)	—
	4					12	20	202	105	63(—,107)	69(47,—)	4.37
	1	33.89	99.80	5		0	4	318	183	135(19,193)	55(36,—)	3.91
	2					4	6	—	—	—(—,—)	—(—,—)	—
51b	1	33.89	99.80	5		0	1	389	148	77(—,125)	80(68,—)	4.27
	1	34.92	100.89	3.5		0	1	144	65	33(—,56)	77(61,—)	4.09
53	2					2	4	—	—	—(—,—)	—(—,—)	—
	1	43.07	86.81	7		0	8	595	427	334(201,48)	44(19,66)	2.50
54	2					8	16	489	354	264(173,40)	46(18,65)	2.46
	3					16	26	522	376	254(177,42)	51(20,67)	2.51
	1	43.08	85.82	25		0	2	36	18	7(—,13)	81(65,—)	4.48
2					2	9	25	15	6(—,10)	75(58,—)	4.15	

Site	Layer	Latitude (N)	Longitude (E)	Average snow depth		Sample depth (cm)		$C_{BC}^{equiv}$ (ng g <sup>-1</sup> )	$C_{BC}^{max}$ (ng g <sup>-1</sup> )	$C_{BC}^{est}$ (ng g <sup>-1</sup> )	$f_{non-BC}^{est}$ (%)	$\dot{A}_{tot}$
				cm	cm	Top	Bottom					
55	3					12	17	111	86	68(25,87)	39(22,78)	2.91
	1	43.51	83.54	25		0	4	146	110	96(52,122)	32(30,63)	2.57
	2					4	8	111	111	97(61,120)	13(7,45)	2.22
56	1	43.66	82.75	24		0	4	90	61	45(16,59)	50(34,82)	2.92
	2					4	8	162	114	99(47,126)	43(27,74)	2.72
	3					8	12	—	—	—(—,—)	—(—,—)	—
57	1	43.64	82.11	15		0	4	151	124	106(57,133)	40(25,58)	2.52
	2					4	9	106	78	62(34,77)	42(27,68)	2.42
	3					9	13	—	—	—(—,—)	—(—,—)	—
58	1	43.52	81.13	37		0	4	37	26	10(1,21)	73(43,97)	3.38
	2					4	8	50	35	18(8,30)	64(38,83)	2.91
	3					8	13	118	83	63(42,85)	47(29,64)	2.25
	4					13	18	61	32	15(5,28)	79(53,—)	3.51
	5					18	23	70	42	25(13,39)	66(43,84)	2.82
	6					25	30	141	133	117(85,147)	17(5,40)	1.96
59	1	44.49	81.15	60		0	5	116	51	24(—,38)	80(68,—)	4.29
	2					5	10	109	37	16(—,27)	85(75,—)	4.41
	3					10	15	195	99	67(15,91)	65(53,98)	3.45
	4					15	20	175	98	73(23,95)	58(46,87)	2.99
	5					20	28	184	97	71(19,93)	62(50,90)	3.08
	6					28	36	204	102	67(—,93)	67(54,—)	3.60
60	1	44.96	82.63	5		0	2	696	542	473(352,58)	32(17,49)	1.84
	2					2	5	—	—	—(—,—)	—(—,—)	—
61	1	44.38	83.09	23		0	3	192	113	83(37,107)	57(45,81)	2.73
	2					3	6	286	197	156(56,200)	45(30,80)	2.88

Site	Layer	Latitude (N)	Longitude (E)	Average snow depth		Sample depth (cm)		$C_{BC}^{equiv}$ (ng g <sup>-1</sup> )	$C_{BC}^{max}$ (ng g <sup>-1</sup> )	$C_{BC}^{est}$ (ng g <sup>-1</sup> )	$f_{non-BC}^{est}$ (%)	$\dot{A}_{tot}$
				cm	cm	Top	Bottom					
62	3					6	15	260	144	110(47,141)	58(46,82)	2.74
	4					15	19	94	69	45(18,60)	52(36,81)	2.85
63	1	44.57	83.96	8		0	1	105	71	50(26,64)	53(38,77)	2.58
	2					1	4	144	80	56(24,74)	61(49,84)	2.79
64	1	45.58	84.29	10		0	1	169	109	82(40,108)	52(37,77)	2.69
	1	46.68	83.54	25		0	2	325	199	152(38,205)	51(34,94)	3.41
65	2					2	6	269	120	84(11,114)	69(58,96)	3.31
	3					6	10	893	363	240(—,336)	73(63,—)	3.75
	4					10	15	268	125	75(—,106)	72(61,—)	3.61
	1	46.49	85.04	4		0	2	156	108	83(33,106)	46(31,79)	2.85
66	1	46.88	85.92	15		0	5	57	40	24(1,33)	58(42,—)	3.48
	2					5	10	134	98	75(22,96)	46(30,85)	3.06
67	1	47.26	86.71	7		0	4	611	446	386(379,48)	38(22,72)	2.73
	2					4	10	298	234	192(96,243)	38(21,71)	2.68
	3					10	16	256	195	140(48,183)	45(28,81)	2.93
68	1	48.15	86.56	32		0	5	140	120	87(46,112)	38(20,67)	2.52
	2					5	10	84	70	46(21,62)	45(26,75)	2.72
	3					10	15	104	82	58(26,77)	44(26,75)	2.74
	4					15	20	20	15	6(1,12)	70(40,95)	3.33
69	1	47.86	86.29	4		0	1	320	264	208(137,26)	36(19,59)	2.22
	2					1	4	191	155	112(64,147)	42(23,67)	2.45
70	1	48.33	87.13	70		0	3	39	24	12(—,18)	68(53,—)	3.93
	2					3	8	31	21	13(0,17)	59(44,99)	3.46
	3					10	15	42	29	18(2,24)	57(42,96)	3.37
	4					15	20	9	6	3(—,4)	68(51,—)	3.69



Site	Layer	Latitude (N)	Longitude (E)	Average snow depth		Sample depth (cm)		$C_{BC}^{equiv}$ (ng g <sup>-1</sup> )	$C_{BC}^{max}$ (ng g <sup>-1</sup> )	$C_{BC}^{est}$ (ng g <sup>-1</sup> )	$f_{non-BC}^{est}$ (%)	$\dot{A}_{tot}$
				cm	cm	Top	Bottom					
71	5	48.07	87.03	40	20	25	18	13	8(1,10)	57(41,94)	3.30	
	6				25	30	29	25	16(7,21)	43(27,75)	2.74	
	7				30	35	21	17	11(3,14)	50(34,85)	3.03	
	8				35	40	36	28	19(6,24)	48(32,83)	2.97	
	1				0	5	96	76	52(21,69)	45(29,78)	2.81	
	2				5	10	88	70	48(20,63)	45(28,78)	2.81	
	3				13	18	30	23	13(2,19)	58(39,92)	3.26	
	4				20	25	33	27	17(7,23)	50(30,80)	2.87	
72	5	47.79	87.56	20	25	30	128	101	85(60,104)	33(18,53)	1.96	
	1				0	3	272	213	135(45,185)	50(32,83)	2.97	
	2				3	8	167	135	90(47,125)	47(26,72)	2.64	
	3				8	12	88	71	40(18,62)	56(30,80)	2.87	
	1				0	4	101	73	50(15,66)	51(35,86)	3.04	
	2				4	9	108	79	56(21,73)	49(33,81)	2.88	
73	3	47.55	88.61	41	10	15	42	30	17(2,24)	61(43,98)	3.43	
	4				15	20	45	34	21(5,28)	55(37,90)	3.16	
	5				20	26	91	73	52(24,66)	43(27,74)	2.69	
	6				26	32	77	63	44(21,57)	43(26,73)	2.67	
	1				0	4	174	139	115(73,142)	34(30,59)	2.24	
	2				4	9	129	95	70(35,90)	46(31,73)	2.59	
74	3	47.63	88.40	34	10	15	76	61	41(17,54)	46(28,77)	2.78	
	4				15	21	73	58	39(16,51)	47(30,78)	2.82	
	5				21	25	83	67	46(21,60)	44(27,75)	2.70	
	1				0	5	45	29	10(—,23)	80(50,—)	3.81	
	2				5	10	38	25	7(—,19)	82(50,—)	3.74	

Site	Layer	Latitude (N)	Longitude (E)	Average snow depth		Sample depth (cm)		$C_{BC}^{equiv}$ (ng g <sup>-1</sup> )	$C_{BC}^{max}$ (ng g <sup>-1</sup> )	$C_{BC}^{est}$ (ng g <sup>-1</sup> )	$f_{non-BC}^{est}$ (%)	$\dot{A}_{tot}$
				cm	cm	Top	Bottom					
76	3					10	15	—	—	—(—,—)	—(—,—)	—
	4					15	22	22	16	4(1,13)	84(40,94)	3.29
	5					24	29	78	53	33(18,49)	58(36,77)	2.66
	6					30	35	45	35	18(8,30)	61(32,81)	2.90
	7					35	40	29	21	7(2,17)	77(39,93)	3.28
	8					40	45	27	19	5(1,15)	81(42,97)	3.40
	1	47.17	88.70	26	2	0	2	315	218	144(77,202)	54(35,76)	2.63
	2				2	2	6	229	168	113(78,159)	51(30,67)	2.30
77	3					6	11	210	173	110(63,158)	48(25,70)	2.56
	4					11	16	93	74	32(15,62)	66(34,84)	2.99
	1	47.27	89.97	45	4	0	4	53	39	17(5,33)	68(39,91)	3.21
	2				5	5	10	40	29	11(3,24)	74(40,92)	3.23
	3				10	10	15	20	14	1(0,11)	97(44,99)	3.46
	4				15	15	20	—	—	—(—,—)	—(—,—)	—
	5				20	25	25	21	16	2(1,12)	91(41,94)	3.29
	6				25	30	30	19	14	1(1,11)	96(44,98)	3.44
78	7					30	35	17	14	1(1,11)	96(40,92)	3.22
	8					35	40	—	—	—(—,—)	—(—,—)	—
	9					40	45	—	—	—(—,—)	—(—,—)	—
	1	46.85	90.32	35	2	0	2	147	75	44(3,63)	71(57,—)	3.73
	2				2	2	7	39	24	10(—,17)	75(56,—)	4.01
	3				7	7	12	23	16	7(—,12)	71(47,—)	3.60
	4				12	17	17	33	22	11(—,18)	67(46,—)	3.55
	5				17	22	17	13	10	4(—,7)	76(46,—)	3.55
6				22	27	22	30	21	11(0,17)	65(44,98)	3.45	

Site	Layer	Latitude (N)	Longitude (E)	Average snow depth		Sample depth (cm)		$C_{BC}^{equiv}$ (ng g <sup>-1</sup> )	$C_{BC}^{max}$ (ng g <sup>-1</sup> )	$C_{BC}^{est}$ (ng g <sup>-1</sup> )	$f_{non-BC}^{est}$ (%)	$\dot{A}_{lot}$						
				cm	cm	Top	Bottom											
79	1	43.53	89.74	30	6	0	78	41	21(—,33)	74(58,—)	4.04							
	2											11	6	62	38	20(—,30)	67(50,—)	3.75
	3											18	11	68	41	21(—,32)	69(53,—)	3.89
	4											24	18	84	50	28(—,40)	67(52,—)	3.71
	5											30	24	83	54	32(4,45)	61(45,95)	3.33
	6											35	30	171	116	86(42,110)	49(35,75)	2.65
	7											40	35	165	99	66(21,87)	60(47,87)	2.98
80	1	44.10	87.49	18	3	0	176	109	67(29,95)	63(47,94)	3.28							
	2											7	3	50	33	12(—,25)	76(51,—)	3.75
81	1	43.60	87.51	6	3	0	172	89	51(8,74)	71(57,—)	3.66							
	2											6	3	189	89	53(1,76)	72(60,—)	3.50
82	1	44.09	84.80	13	5	0	48	24	9(—,15)	81(68,—)	4.66							
	2											9	5	28	16	8(—,11)	73(59,—)	4.22
83	1	43.93	85.41	12	3	0	101	66	37(3,53)	64(48,—)	3.54							
	2											7	3	48	31	14(—,23)	71(52,—)	3.81
	3											12	8	1087	841	619(336,76)	43(29,69)	2.44
84	1	43.93	86.76	25	2	0	489	372	250(127,34)	49(30,74)	2.63							
	2											6	2	76	50	7(—,38)	91(49,—)	3.68

**Table 32.** Surface and subsurface snow sample average values within the five sample regions in Figure 1.

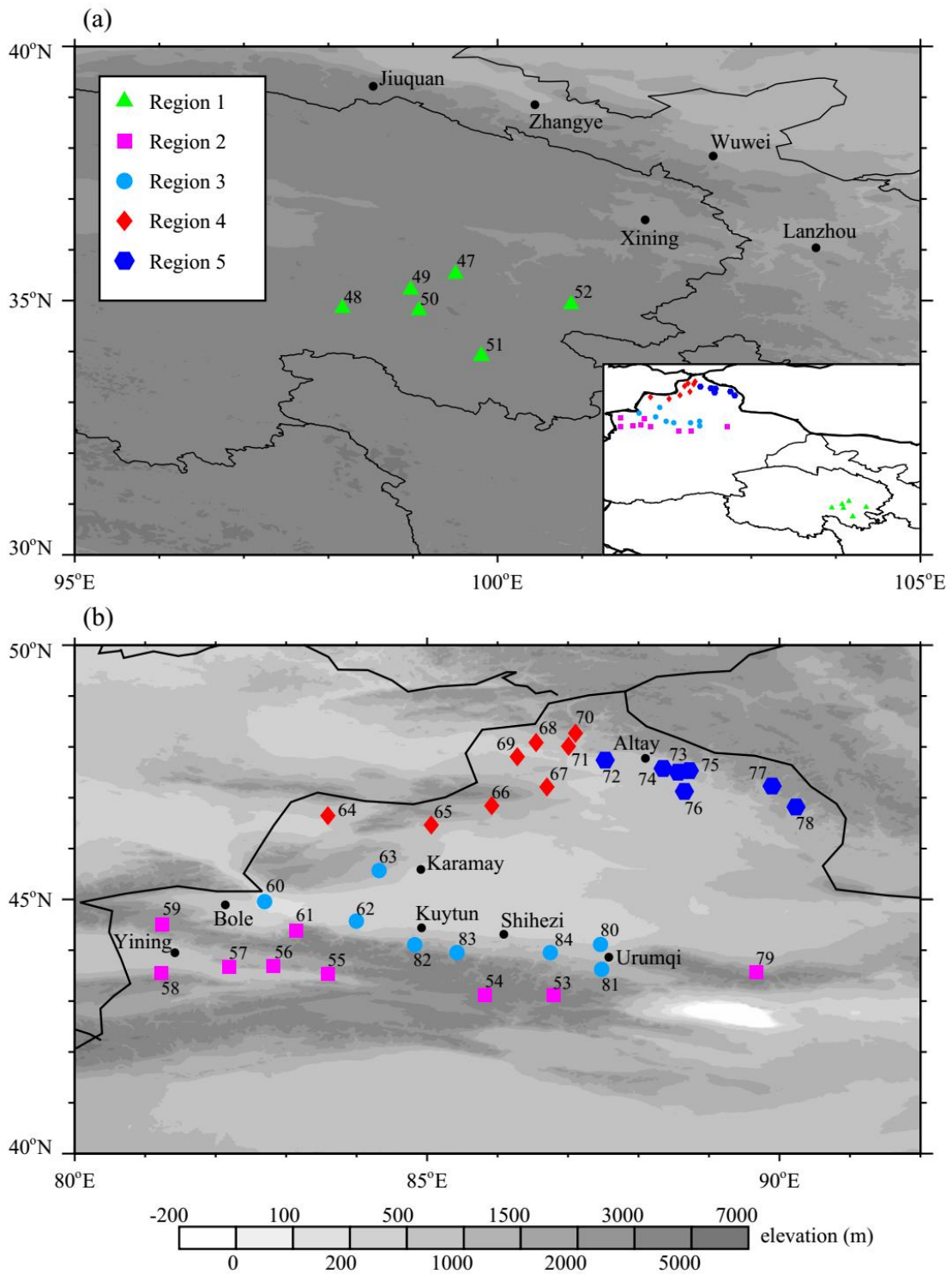
Regions	Layers	$C_{BC}^{\max}$ (ng g <sup>-1</sup> )	$C_{BC}^{\text{est}}$ (ng g <sup>-1</sup> )	$C_{BC}^{\text{equiv}}$ (ng g <sup>-1</sup> )	$\dot{A}_{\text{tot}}$	$f_{\text{non-BC}}^{\text{est}}$ (%)
1	surface	151±66	99±53	307±119	4.24±0.29	70±11
	subsurface	174±110	113±78	332±201	4.17±0.19	66±4
2	surface	108±126	81±102	160±171	3.27±0.80	59±19
	subsurface	119±96	89±69	170±134	2.86±0.49	49±16
3	surface	173±183	127±158	245±226	3.11±0.86	58±15
	subsurface	117±158	75±120	175±201	3.51±0.50	72±12
4	surface	160±141	126±124	218±192	2.99±0.56	48±11
	subsurface	114±77	82±56	178±157	2.96±0.36	51±10
5	surface	112±79	74±56	158±104	3.09±0.56	58±16
	subsurface	61±46	37±31	80±57	3.03±0.37	62±16

**Table 43.** Total snow water equivalent (SWE) and estimated total snowpack BC mass in a 1-cm<sup>2</sup> column of snow.

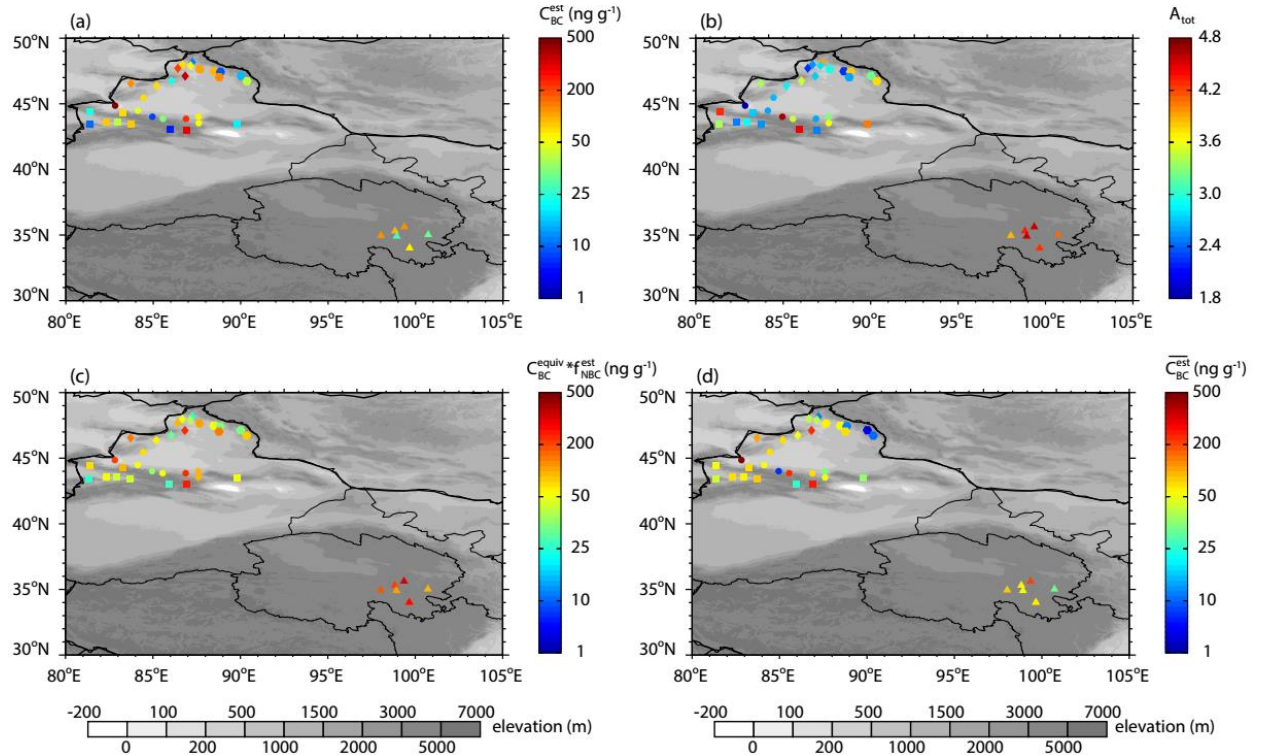
Site	Date sampled (2012)	Integrated SWE (g cm <sup>-2</sup> )	Integrated BC (ng cm <sup>-2</sup> )	Snowpack average
				BC mixing ratio (ng g <sup>-1</sup> )
47	10 Jan	5.94	1212	204
48	11 Jan	0.51	54	106
49	11 Jan	3.77	—	74
50	1 Jan	1.56	—	57
51a	12 Jan	1.52	—	135
51b	12 Jan	0.14	11	77
52	13 Jan	0.14	—	33
53	31 Jan	9.18	2632	287
54	1 Feb	2.41	64	27
55	2 Feb	1.16	112	96
56	2 Feb	1.16	—	76
57	2 Feb	1.99	—	80
58	3 Feb	5.33	244	46
59	4 Feb	7.67	451	59
60	4 Feb	0.22	—	473
61	5 Feb	2.87	265	92
62	8 Feb	0.56	30	54
63	9 Feb	—	—	—
64	9 Feb	2.2	283	129
65	10 Feb	—	—	—
66	10 Feb	1.45	80	55
67	11 Feb	5.64	1320	234
68	11 Feb	2.9	128	44
69	11 Feb	—	—	—
70	12 Feb	7.9	99	13
71	12 Feb	5.15	226	44
72	14 Feb	1.74	138	80
73	14 Feb	5.96	236	40
74	14 Feb	4.16	235	56
75	17 Feb	7.64	—	12
76	18 Feb	2.68	230	86
77	19 Feb	6.1	—	4
78	20 Feb	5.07	53	10
79	21 Feb	7.05	282	40
80	23 Feb	0.91	32	35
81	23 Feb	1.02	53	52
82	24 Feb	0.97	8	8
83	24 Feb	1.43	343	240
84	25 Feb	0.66	58	88

**Table 54.** Average regional contributions to the 650-700-nm particulate absorption.

Regions	Industrial pollution (%)	Biomass burning (%)	Soil dust (%)
1	12	59	29
2	25	60	15
3	58	31	10
4	27	67	5
5	41	28	31

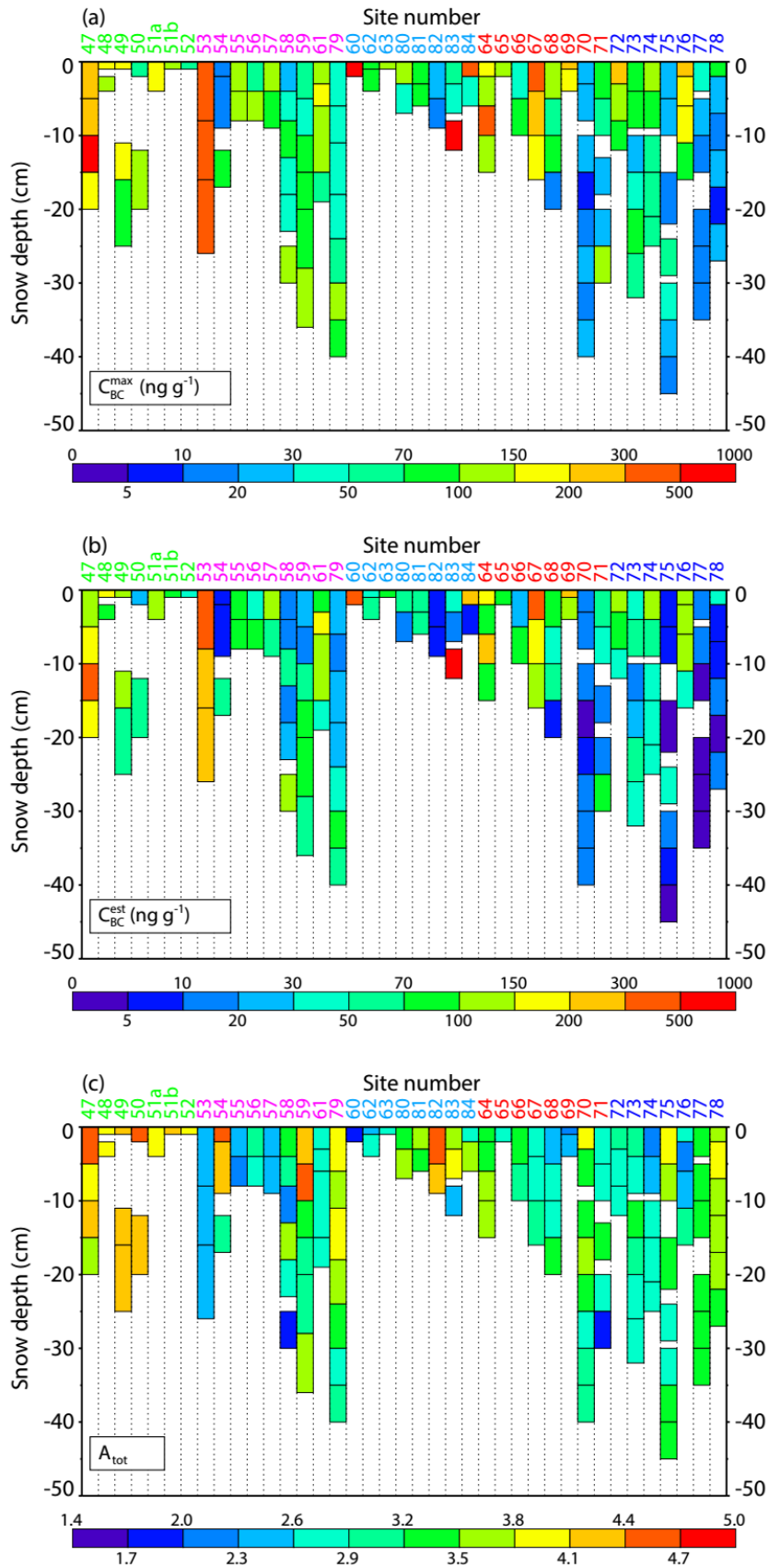


**Fig. 1.** Snow sampling locations, site numbers, and regional groupings in (a) Qinghai and (b) Xinjiang, black filled circle are the locations of main cities on the map.

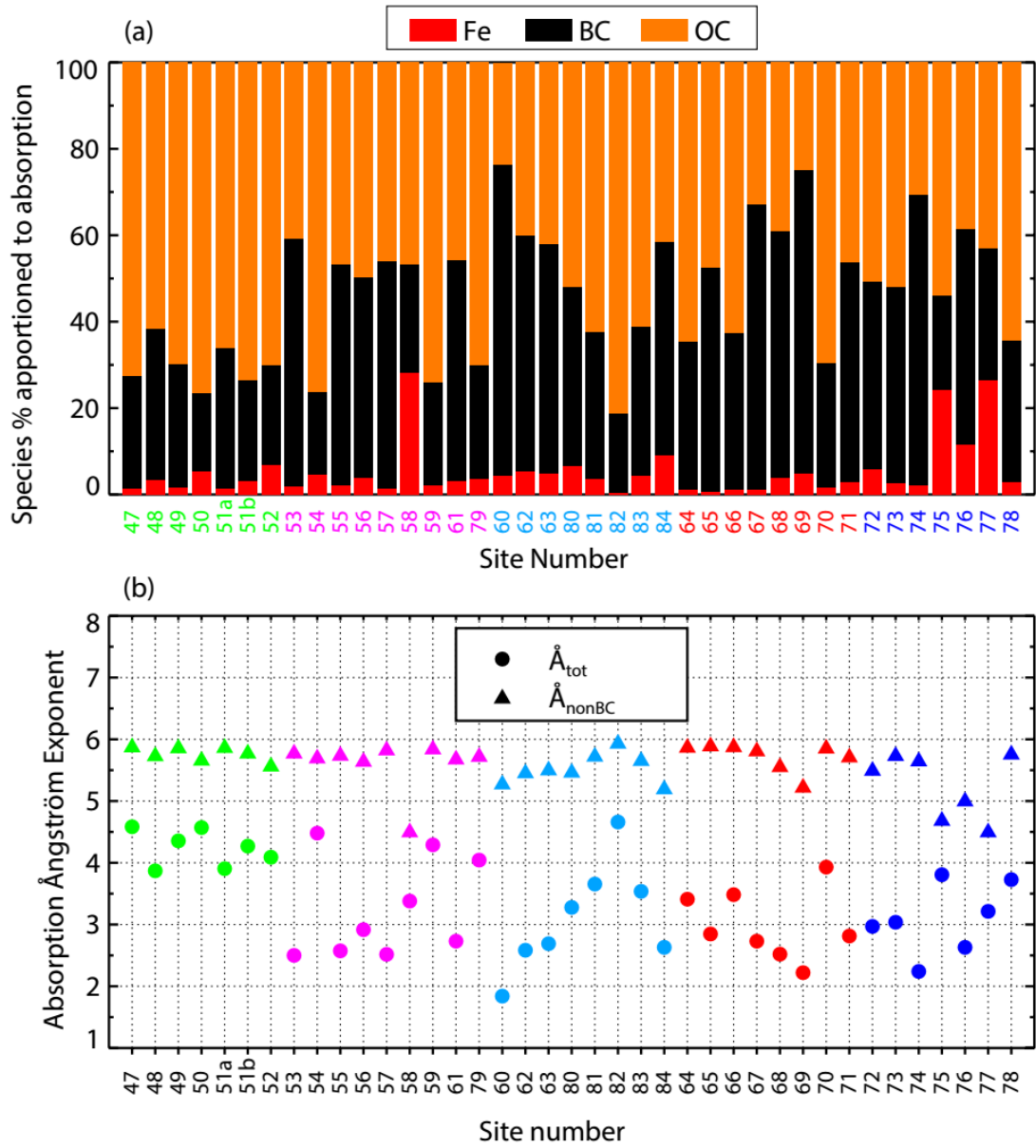


**Fig. 2.** (a)  $C_{BC}^{est}$  and (b)  $A_{tot}$  for the surface layer at each site, with symbols according to region (see Figure 1). (c) Estimated BC-equivalent mixing ratio (ng g<sup>-1</sup>) required to explain the spectrally integrated (300–750 nm) absorption of sunlight by non-BC components in snow. (d) Estimated average snow BC mixing ratio,  $\overline{C_{BC}^{est}}$ , which was calculated by integrating the snow water content and BC mass over the entire snowpack (see Table 43).

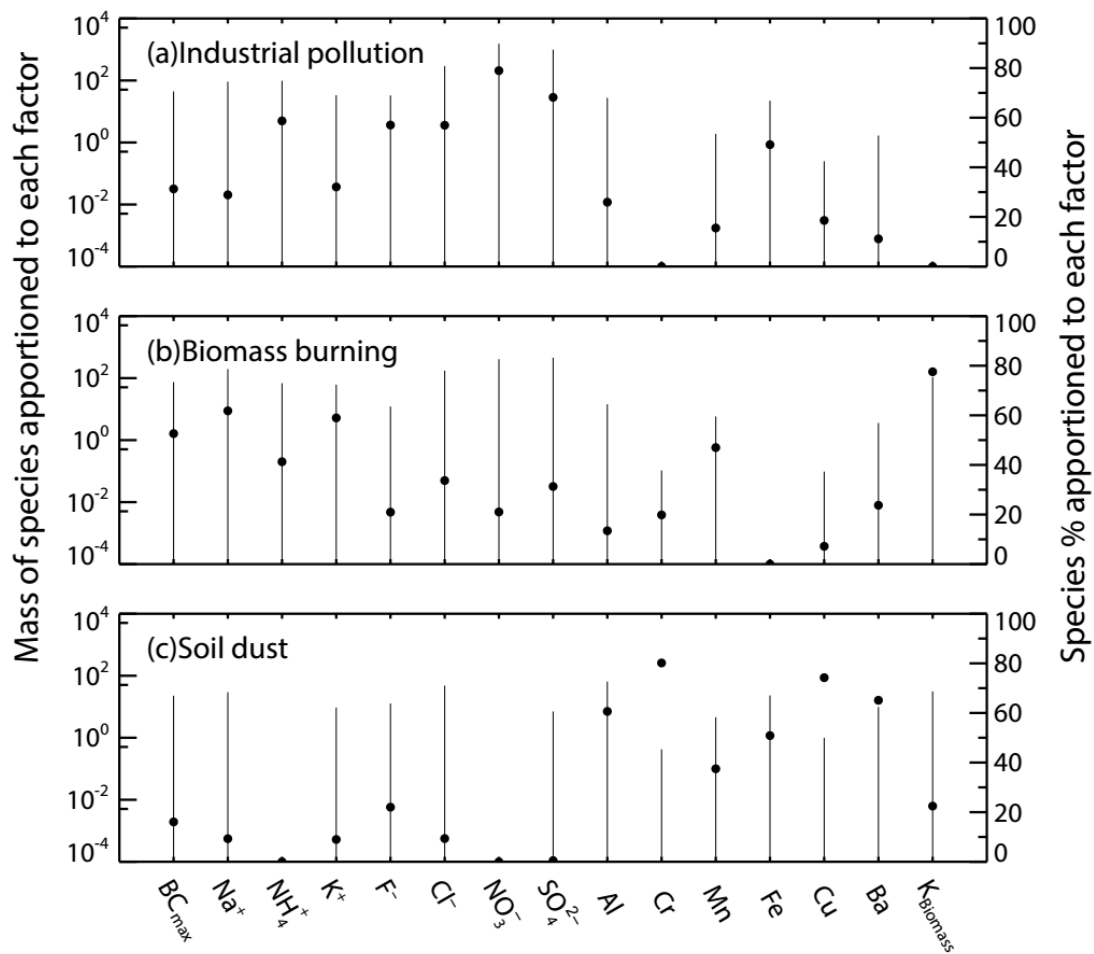




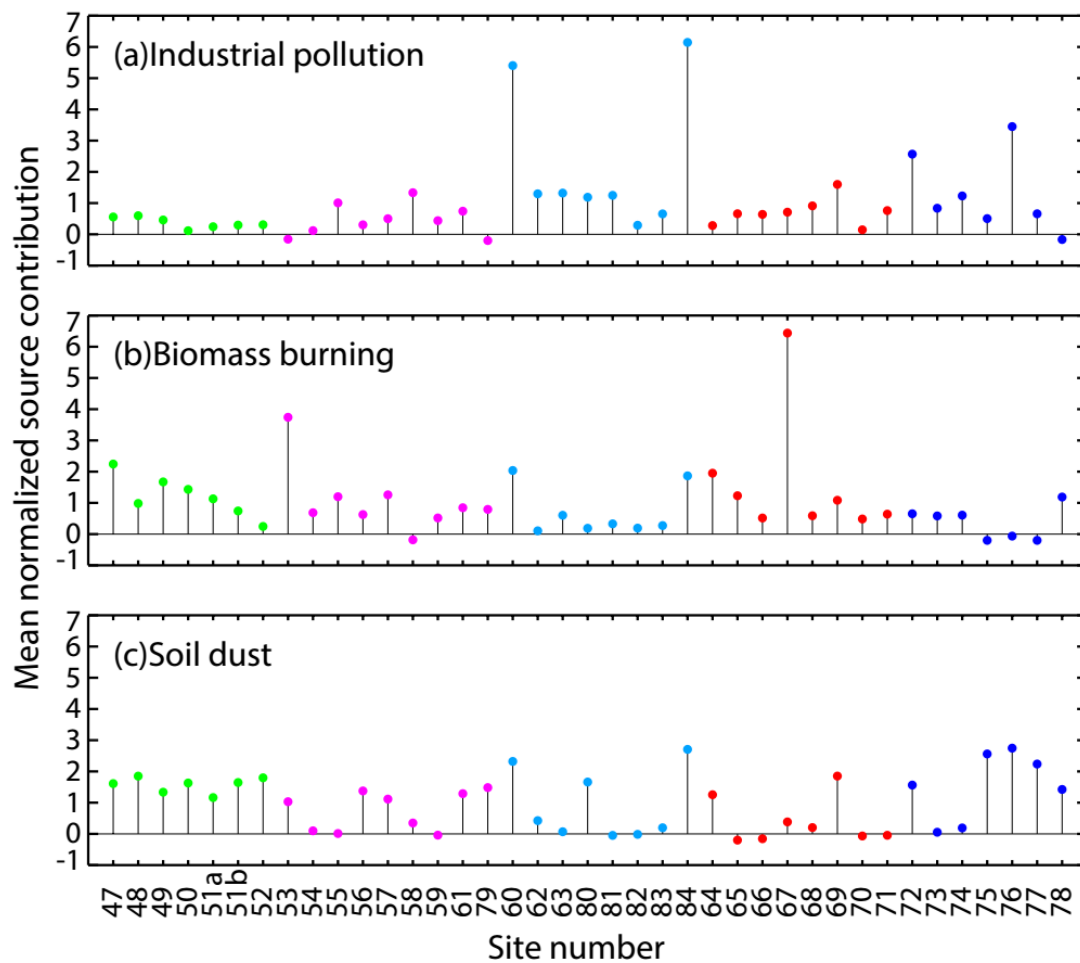
**Fig. 3.** Vertical profiles of the (a)  $C_{BC}^{max}$ , (b)  $C_{BC}^{est}$ , and (c)  $A_{tot}$  at each sampling site.



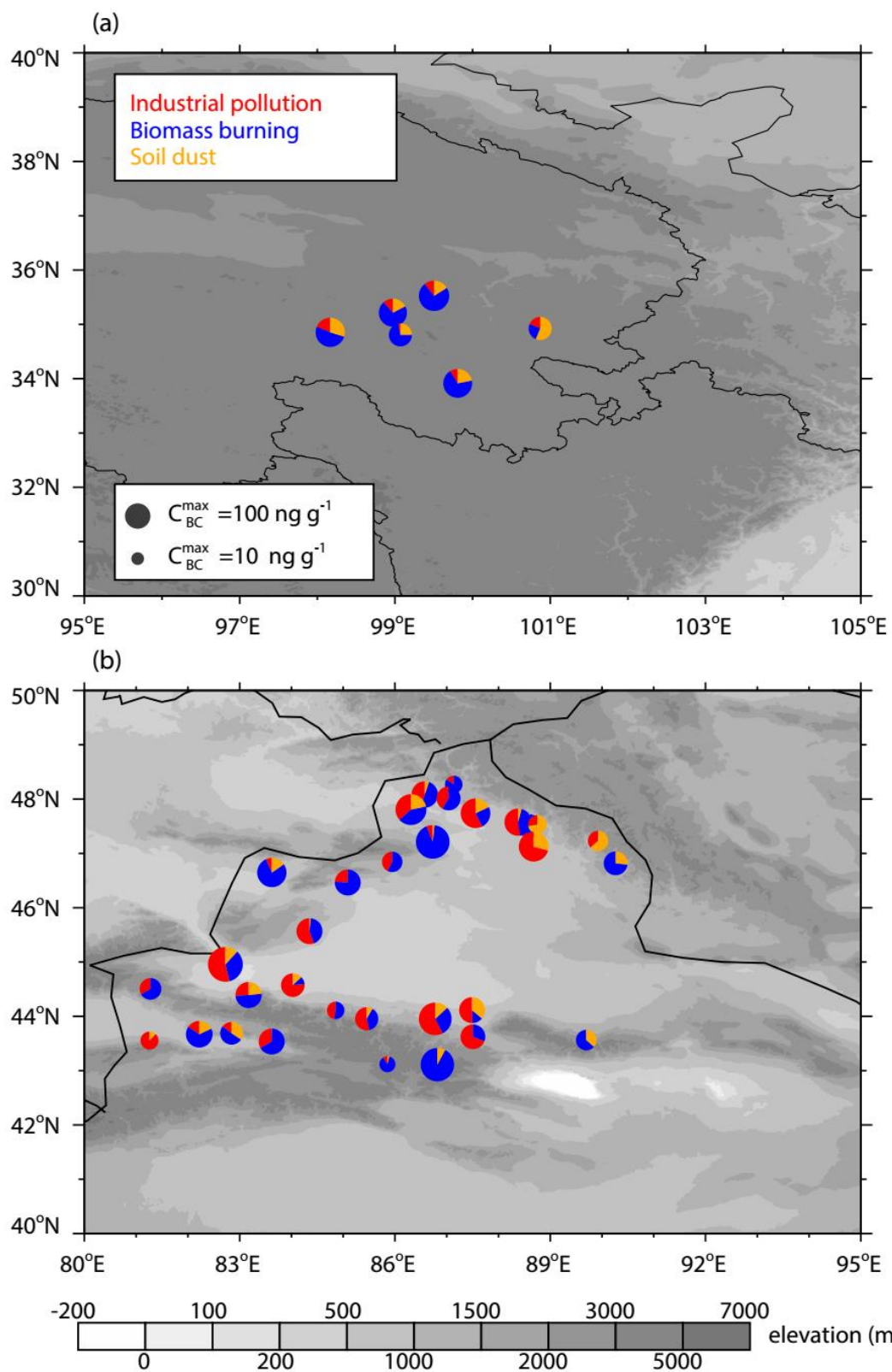
**Fig. 4.** (a) Relative contributions from the BC, OC, and Fe oxides to the total absorption optical depth for the surface snow samples. (b)  $\hat{A}_{tot}$  and  $\hat{A}_{nonBC}$  for the surface snow samples.



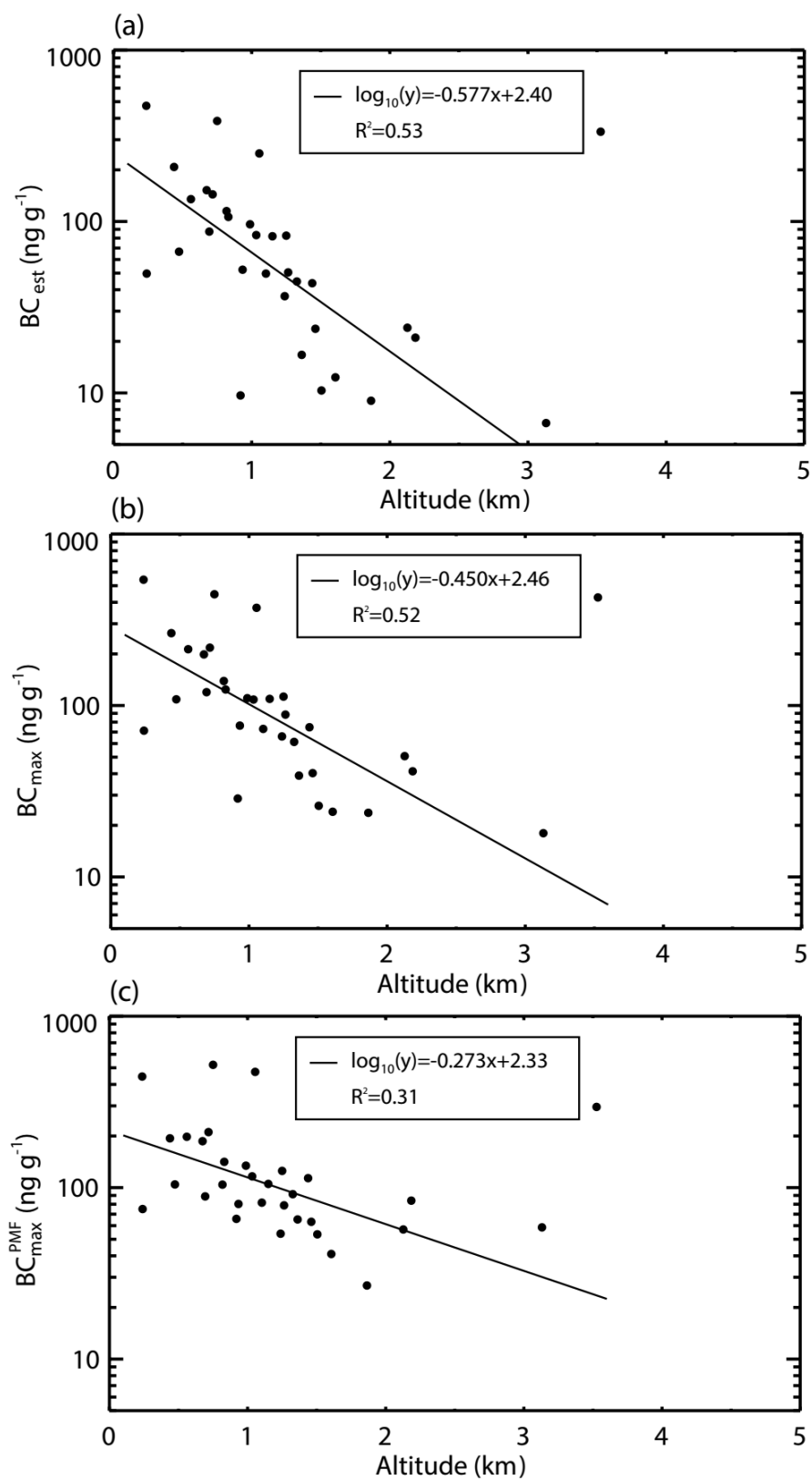
**Fig. 5.** Source profiles including the measured mass concentrations (lines) and the percent of [componentsspecies](#) (dots) for the three factors/sources that were resolved by the PMF 5.0 model.



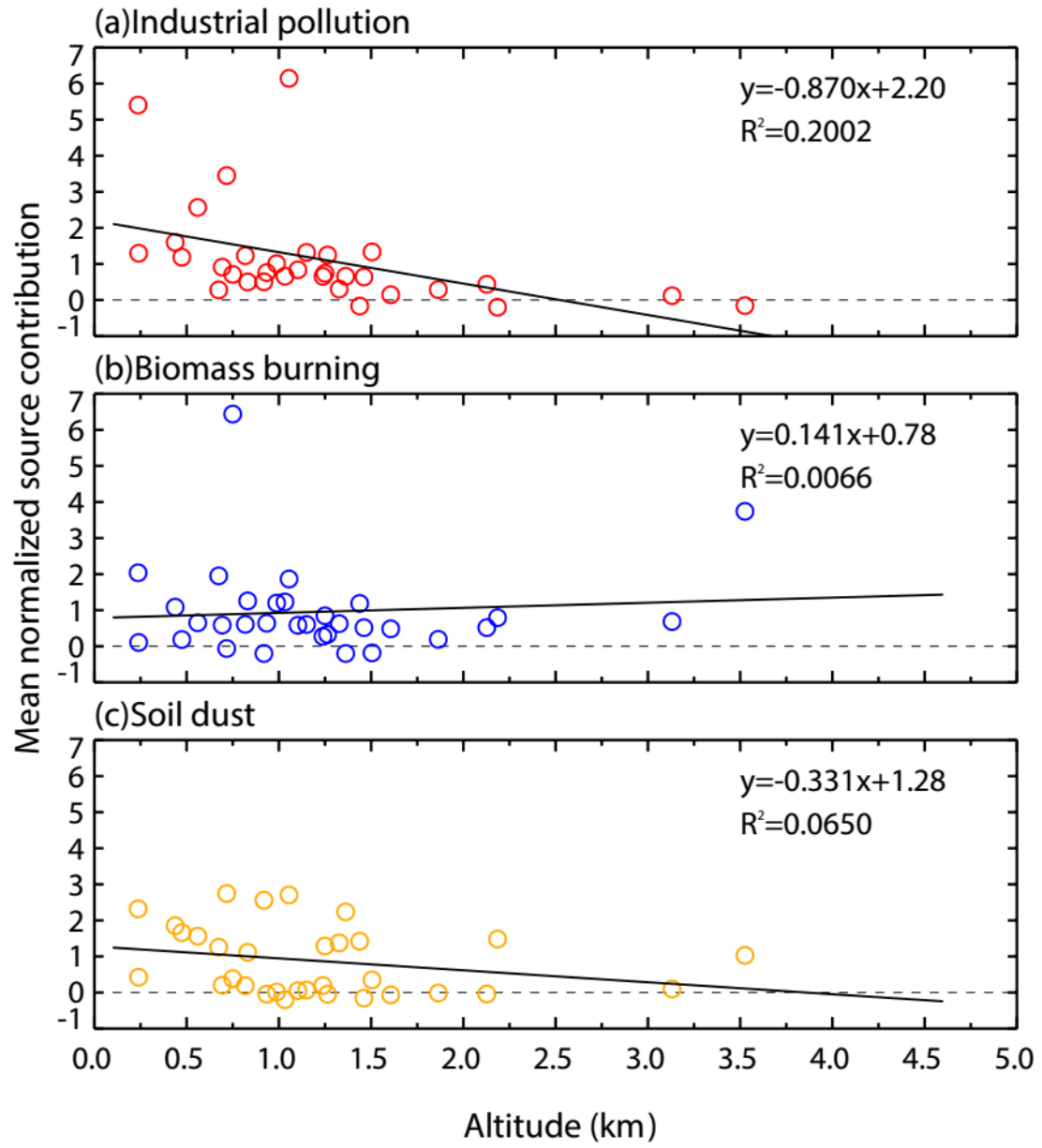
**Fig. 6.** Contributions from each source/factor to each sample or individual receptor site. The contributions have been normalized by the average value of the respective factor contributions over all sites.

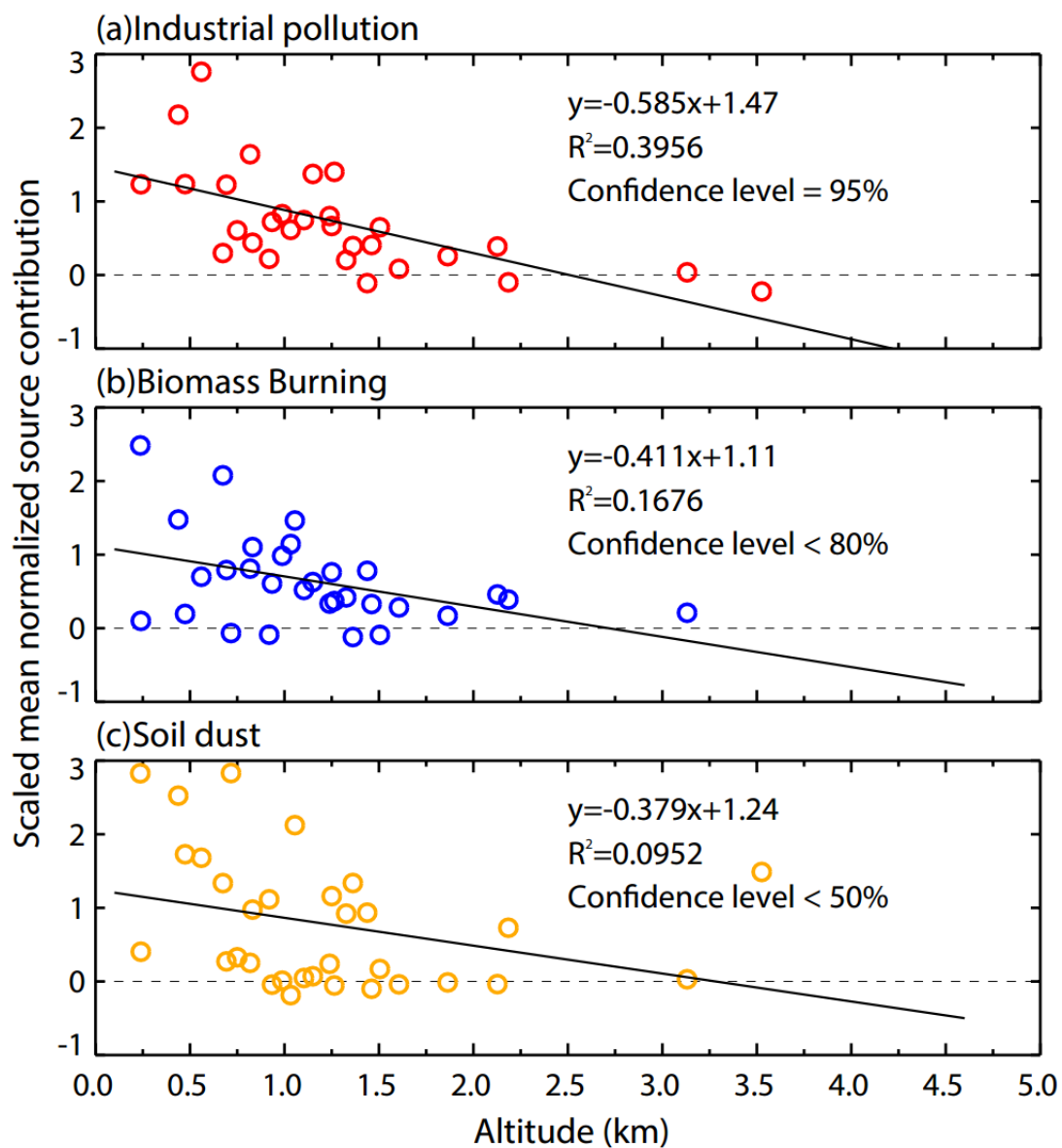


**Fig. 7.** Fractional contributions from soil dust, biomass burning, and industrial pollution to 650–700-nm particulate absorption according to the PMF analysis for the surface snow samples in (a) Qinghai and (b) Xinjiang.



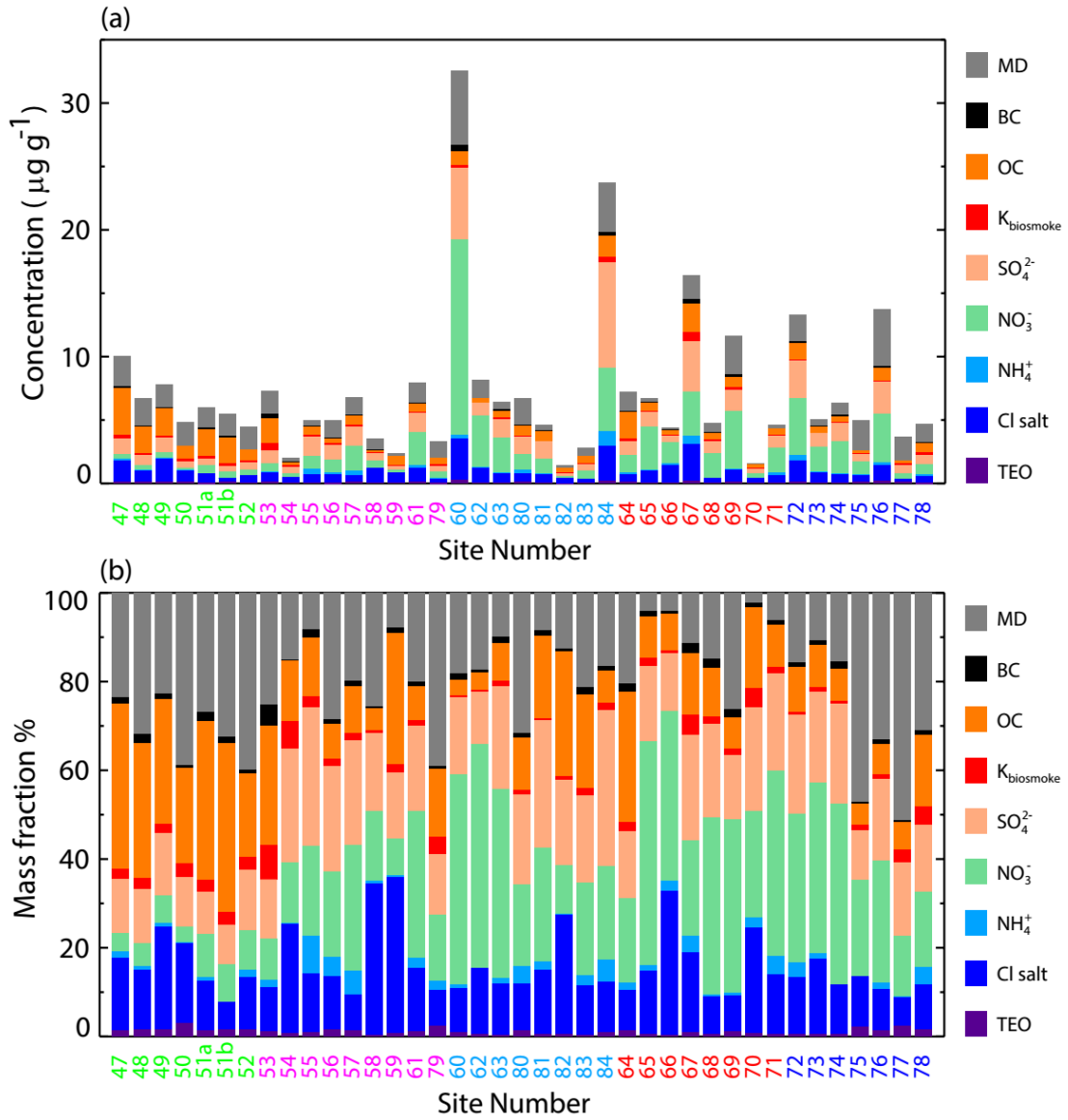
**Fig. 8.** (a)  $C_{BC}^{est}$  and (b)  $C_{BC}^{max}$  measured according to the ISSW and (c)  $C_{BC}^{max}$  determined by the PMF model for the surface snow for each site as a function of altitude for the sites in Xinjiang.



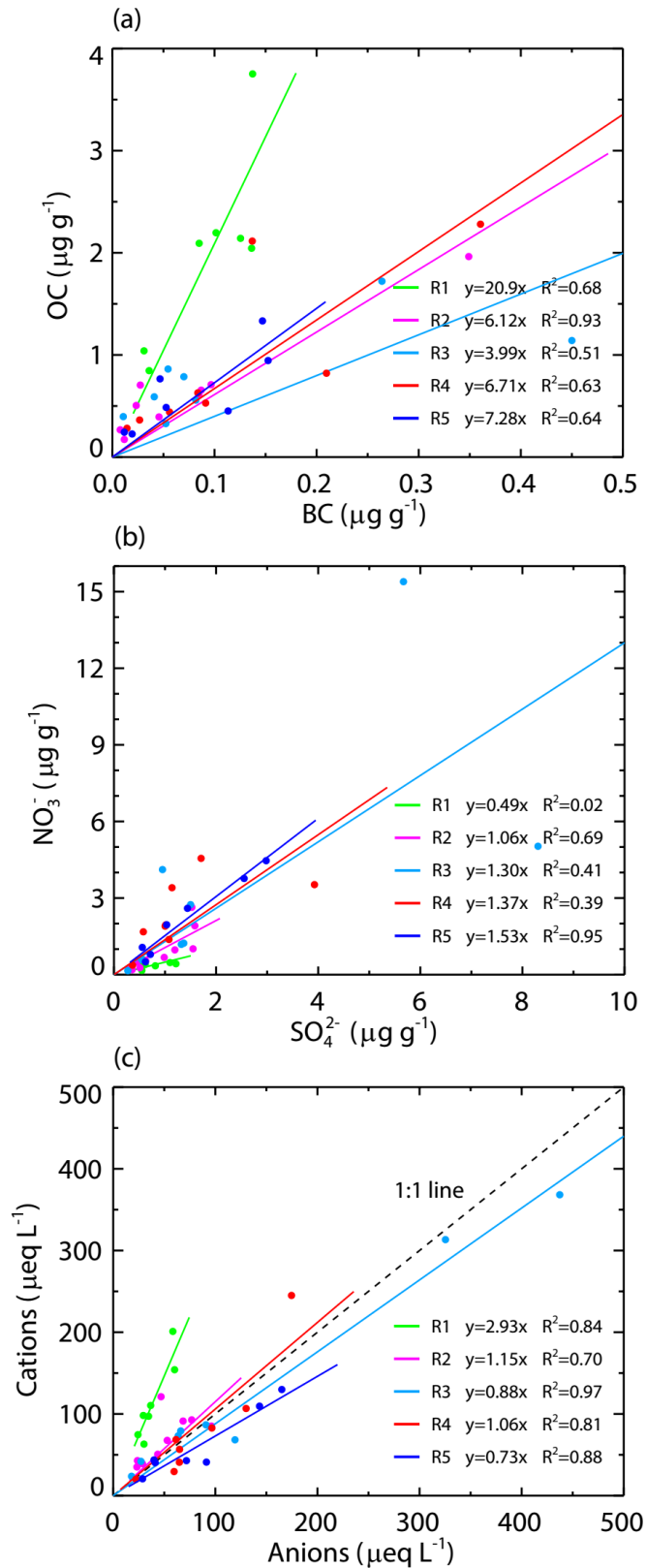


**Fig. 9.** Scaled Contributions from each source/factor as a function of altitude at sampling sites for the sites in Xinjiang. The contributions were normalized by the average value of the respective factor contribution over all sites.





**Fig. 10.** (a) Average mass contributions and (b) average mass fractional contributions of the chemical components in the surface snow at each site.



**Fig. 11.** Ratios of the (a) OC and BC mixing ratios and (b)  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$  concentrations, (c) and the charge balance between cations ( $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$  and  $\text{NH}_4^+$ ) and anions ( $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ ,  $\text{Cl}^-$  and  $\text{F}^-$ ) for the surface snow samples.

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