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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Source attribution of light-absorbing impurities in seasonal snow across northern China" *by* R. Zhang et al.

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General comment. We thank both reviewers for their comments and employ the same numeration they do when responding to their specific comments. We also note that, to be consistent with revisions made to nomenclature in companion papers after this study was submitted, we now refer to LAI as Insoluble Light Absorbing Particles (ILAP).

Reviewer No. 1. Major Points 1.1 The relationship of this study to that of Wang et al (2013) and Huang et al (2011). The reviewer wishes us to differentiate the work presented here from that of Wang et al and Huang et al. The Huang et al paper in the Bulletin of the AMS was essentially a report of the occurrence of the study, the justification for such a study, and a report of some preliminary data to highlight the potential value of such an analysis. The data reported were visual estimates, in

the field, of the light absorption present in the snow melt filters. These data were superseded by the quantitative spectrophotometric data reported in Wang et al. Hence, in our view, the differentiation of importance is that between the current study and that of Wang et al. The Wang et al study reported spectrophotometric measurements of the absorption of insoluble light-absorbing aerosols as a function of wavelength. From this, the total light absorption by all insoluble (filterable) particles (ILAP) in the snow samples could be determined with considerable accuracy. Further, using various assumptions as to the absorption Ångstrom coefficient of non-BC articulate light absorbers in the snow, a rough and rather uncertain estimate was made of the relative contributions of BC and non-BC aerosol species. This information, coupled with values for iron (Fe) concentrations in the snow aerosol (an indicator of the absorption by mineral dust) was used to give estimates of the relative contributions of iron oxides, organics and BC to the aerosol light absorption in the snow for the various regions sampled in the study (i.e. Wang et al. Figure 11). However: 1. these estimates rely on very uncertain assumptions about the spectral absorption properties of these three categories of light-absorbers, and so themselves are very uncertain, and 2. these estimates separated contributions by category of light-absorber, not by source type. The latter is a critical distinction, because, for example, light-absorbing organics could come from soil or from combustion (so-called "brown carbon"); the optical analysis of Wang et al. cannot distinguish the two. Similarly, while iron oxides are expected to mostly be associated with mineral dust, they may also come from industrial sources.

The current study uses only the measurements of the total light absorption by ILAP from the Wang et al study. It then uses the concentrations (directly measured) of 14 other chemical species (with the exception of Fe, not reported in Wang et al) in the snow aerosol in a PMF analysis to yield quantitative measures of the contributions of the identified source profiles to the light absorption by ILAP in the snow. In other words, the light absorption is associated with specific sources, rather than with categories of species (as is given by Wang et al.), and it does not rely on very uncertain assumptions about aerosol species' optical properties. Additionally, to support the PMF analysis, a

back trajectory analysis was performed to see how consistent the PMF sources were with the geographic distribution of known ILAP sources and the winds prevalent during the snow deposition period. These analyses are quite distinct from those done in Wang et al, which contains neither quantitative source attribution nor trajectory analysis. The above discussion is of course much too lengthy to be included in the text. Nevertheless, while we feel that the differences between this work and that presented in Wang et al is already clear, we have added some discussion to the second paragraph of the introduction to further clarify the distinction. There we note that an appraisal of the relative contributions of BC, OC and Fe oxides to light absorption in the snow was made in Wang et al. As to mentioning the Wang et al study in our discussion, we do this more than a dozen times in the current text and see no need for further and redundant citation. 1.2 The use of BCmax rather than other parameters such as BCest. As we state in the text (Section 2.2), we selected BCmax because it is the most unambiguous measurement available. We also chose it because it quantifies the absorption by ALL ILSP, not simply BC. It does this by taking the absorption and dividing it by the mass absorption crossection of the calibration aerosol – which is a form of BC (see Doherty et al, 2010 and Grenfell et al, 2011 as cited in the text). It thus gives the amount of calibration aerosol that would be necessary to yield the observed total absorption in the 650-750 nm band. It is really a scaled total absorption by all ILAP and NOT the absorption due to BC alone (which would be BCest and, as the name indicates, can only be estimated with various assumptions). Remember that the differentiation between the various ILAP components (e.g., BC, soil dust, mineral dust, etc.) in Wang et al is based largely on optical measurements and various assumptions about the optical properties of the prospective ILAP components – hence the need to calculate these other parameters. In our case, the allocation of absorption amongst the various ILAP components is based on the chemistry of co-deposited chemical species and a PMF analysis. We have no need to differentiate between the components and in fact it would be dysfunctional to do so. We will add to the discussion in the first paragraph of Section 2.2 to emphasize that the parameter BCmax is a scaled absorption by ALL

ILAP.

1.3 Contrast in the allocation of total absorption by ILAP between Wang et al and this study. The reviewer cites Figure 11 of Wang et al, which shows a very modest contribution from Fe absorption to the total ILAP absorption. On this basis he asserts that there is a contradiction between the results presented here and the Wang et al analysis. While there is indeed some contrast in our conclusions and those of Wang et al, there is no real contradiction. What Figure 11 shows is that there is a very modest impact of the mineral (iron oxide) component of dust on the total absorption. What we are discussing is SOIL, of which mineral dust is a variable and commonly small component. There are other species in soil dust than iron that absorb visible light, such as organic carbon. As can be seen in Figure 11 of Wang et al., OC makes a significant contribution to light absorption, and as noted above and in Wang et al. this OC could be from either soil or combustion aerosol. We feel we can do no better than to cite the discussion in Wang et al on this point. From the introduction in Wang et al (3rd paragraph), “Organics in the form of humic-like substances (HULIS) in soil also absorb solar radiation. . . snow albedo in this region may be strongly affected not only by BC but also by combustion organics, soil organics and iron oxides in soil and mineral dust.” Furthermore, our source types and those of Wang et al as shown in Figure 11 are not equivalent. Wang et al deal with BC, OC and Fe absorption. These are clearly not sources but absorbing species (or groups of species). We, on the other hand, identify actual sources, namely biomass burning and biofuels, industrial/urban aerosol and soil dust. These sources can each have multiple absorbing species in them. For example, biomass burning and biofuels will clearly contain BC and OC, industrial/urban aerosols will contain BC, OC and quite likely some Fe, and soil dust will contain OC and Fe. Given this, we see no contradiction between our results and those of Wang et al. For example, in the Northeastern region (our regions 5 and 6 – which correspond roughly to region 4 of Wang et al), Wang et al see a predominant impact of BC. We see that biomass burning and biofuels, and industrial/urban aerosols are together the largest component of the ILAP – and they contain virtually all of the BC present. Hence, while

Wang et al would attribute $\sim$ 80% of the absorption in this region to BC, our PMF results would suggest a lower figure, perhaps 50-60%. Given the rather high uncertainty in the BC/non BC partitioning implicit in the approach of Wang et al (Doherty et al, Atmos. Chem. Phys., 10, 11647-11680, 2010), we feel that the disagreement between our conclusions and those of Wang et al are rather mild. Nevertheless, we will insert text in section 3.1 and the Conclusions to put our results in context with the earlier work of Wang et al.

1.4 Back trajectory analysis, number of trajectories. For each region, as stated in the text, a representative site was selected and anywhere from $\sim$ 300 to 500 trajectories were run over the deposition period for each selected site. We now state this in the text.

1.5 Observed precipitation characteristics from the CMS. The reviewer suggests that we make use of the temperature as well as precipitation data from the CMS in our assessments of the amount of snow deposited at each site. We in fact did this, together with any observations of the precipitation type. We now state this in the text and give the URL for the CMS data.

1.6 Local dust and filtering of snow samples While the reviewer raises an interesting issue here, we again must disagree somewhat with the utility of his proposed alteration to the sampling protocol. (In any case it is a moot point – the sampling was completed several years ago. When the sampling was undertaken, it was not at all clear that local soil dust would be such a large part of deposited aerosol.) Several points are relevant to this discussion. First, sampling with filters at reasonable cut points would have divided the sample, and absorption, over multiple filters, thus lowering quite significantly our signal to noise ratio. Second, the snow samples, after melting, were commonly allowed to settle briefly before filtration, thus eliminating some of the most highly local dust. The impact of these course local particles on the snow albedo has in fact been estimated in Wang et al based on this sedimentation. Third, it is not the intent of our analysis to differentiate between local and distant dust source but rather between all dust sources

and chemically distinct sources such as biomass burning. The discussion of local soil dust is simply an appraisal of why some of the samples in regions 5and 6 have unexpectedly high dust factor contributions.

Minor Points

1.1 The text on line 16 has been amended to read: "...in China including rapidly increasing industrial/urban emissions."

1.2 We have substituted Warren and Wiscombe for Warren as suggested by the reviewer.

1.3 The text has been altered to read: "...ILAP in Chinese snow...."

1.4 Zhang et al give two URL's from which gridded emissions data can be downloaded. This data was then plotted with the TrajStat software package (Wang et al, 2009, as cited in the text). We note this now in the text at this point, in addition to a later citation already in the text.

1.5 Nowhere do we claim to separate BC from other ILAP, certainly not here. On the contrary , it is because BC is not separated from the other ILAP that we use the parameter BCmax and do source allocation via PMF.

1.6 We have explicitly named the two major corrections already and feel that the detailed discussion in Wang et al (2013) is easily accessible and preferable to a redundant exposition here.

1.7 The US EPA PMF 4.1 model is not available to the general public. We are essentially Beta testing the model. That is why we do not report the more sophisticated error analysis of the 4.1 model but confine ourselves to precisely the same analysis as is available from the 3.0 model. This latter model is in fact available at the EPA web page: http://www.epa.gov/heasd/products/pmf/pmf.html

1.8 Both are acceptable but the reviewer is correct in that "Chinese" would be preferred

and we have made this change.

1.9 We feel that the addition of these figures to the main text would break up the text and interrupt the flow of the discussion more than is necessary.

1.10 The data for the generation of the arid regions shown in Figure 1 (and the other figures) were included with the TrajStat software downloaded from the site: http://www.meteothinker.com/TrajStatProduct.aspx. They have been used in a number of previous studies (cf., Gong et al, J. Geophys. Res., 108, doi: 10.1029/2002JD002633, 2003).

1.11 We presume that the reviewer is referring to the use of LAI (it is not clear). If so, we will now use ILAP as per our opening remarks.

1.12 The time periods for each region are in fact different and we now cite them in the text.

1.13 We are not sure how to address this point since no specific instances are given. In our view, the English usage is generally clear.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 2155, 2013.