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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Single particle characterization of black carbon aerosol in the Northeast Tibetan Plateau, China" *by* Q. Y. Wang et al.

Anonymous Referee #1

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The dataset as provided by this manuscript is scientifically important because of the unique location of the experimental site, the direct measurements of BC particles by the SP2, in addition to the rarity of such kind of ground based dataset sampled from Tibetan plateau. This is a potentially important manuscript according to the instrumentations used, local/regional pollution influences and meteorological conditions as mentioned by the authors, however, this paper may have been submitted too rush and the data analysis has not been fully completed. There are many places where the conclusions are not solid and lack of sufficient data analysis support; many of the discussions are superficial and surely need to be revisited. I think this paper needs to be more carefully written and I would suggest the authors to complete a comprehensive data analysis

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and careful writings before the publication can be considered. The major issues are listed below.

1. About the MAC calculation and the interpretation of Aeth data. First of all, this part of abstract is not quite pleasant to read: the abstract line 15-18, after you have just stated that "the Aeth data is not reliable", how could you use Aeth to derive MAC? I kind of understand what you mean, the Aeth is not suitable for BC mass determination but only used for absorption measurement (even the absorption data you have used may be wrong as well), but please state these more carefully and logically in the abstract.

For the Aeth data interpretation, you used an attenuation coefficient (σ) which is manufacture defined, to derive the Aeth BC mass. Firstly, this value could be wrong, we can't just use the same value under different atmospheric conditions, because the MAC of BC will be influenced by many factors in the real atmosphere as you have stated, but you have just made a self-conflict after you have made these statements: in Fig. 7A the Aeth BC mass is compared with SP2 BC mass, this will absolutely confuse the reader as the Aeth BC itself is wrong. It is not a good idea to use anything about Aeth BC given you are talking about the MAC in this paper, the better approach for Fig. 7 is to plot the Aeth absorption VS SP2 rBC, then apply for a linear fitting to get the least square fitted MAC (=absorption/mass). And definitely you should discuss in more details where the difference of MAC may result from, between the urban environment Xi'an and this site QHL, but you haven't explained these at all, just simply listed the values from other literatures.

Secondly, it seems that the Aeth data has not been interpreted properly. You claimed "BC concentrations there derived from Aethalometers may also need careful examinations and possibly new interpretations". Please do it, please do not just use the directly measured attenuation from Aeth, the non-absorbing particles will surely contribute to the total light extinction/attenuation, there is no doubt that this will overestimate the "absorption". We should at least correct for the aerosol scattering properties (There are so many literatures discussing this correction I am not going to list here). You have just

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duplicated claiming this truth at many places but have not corrected for it. Given the MAC value is one of the main conclusions, we can't use this extinction-derived "MAC" and even put this wrong MAC into the abstract.

2. The SP2 data has not been fully analysed. Have you done the LEO fitting for the scattering signal of BC? We can't use the raw scattering signal of BC as in Fig.2. I know the approach in Fig. 2 is from Schwarz's paper 2006, but that is one of the earliest SP2 papers, there are so many updates since then, we are not currently only relying on that plot to derive the "internally mixed" fraction. About the SP2 calibration, you used fullerene, please also state how you get the single particle mass from modality diameter.

In Fig.2, you set an arbitrary threshold of delay time above which the incand/scatter signal looks large variation however the delay time varies little, but it has not been physically clearly explained why this arbitrary delay time value should be selected. From the image coloured by the single particle mass, it can be seen that the above 2us delay time is mainly composed if smaller particles, it means by this method, you have just selected the smaller BC particle to be "internally mixed" whereas the majority of the larger particles are externally mixed? It is not true in the real atmosphere. More importantly, would you like to explain more precisely your definition of "internally" or "externally" mixing?-Chemically based or size/physically based? Again, please refer to the recent methodology to determine the "thickly coated" fraction from SP2 measurements.

3. About the BC mass loading the conclusion in page 21953: "These comparisons suggest that the values found at the rural QHL site are well above background values and, hence, are very likely influenced by local rBC sources."1) There is no point to compare this plateau environment (over 3000m a.s.l.) with the urban studies, the urban is certainly more polluted; 2) You compared the SP2 results with the Aeth data from many other sites, it is making large confusion to compare the BC mass from different instruments(given you have got Aeth BC as well, why not directly compare using the same instrument); 3) It may be worthwhile to compare the BC mass at this

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plateau site with the other measurements which were also conducted at a high altitude station, but you should list a table clearly stating the altitudes, instrumentation, experimental periods(seasons), local influence at al.. You have not stated anything about the measurements from the other sites as you have referenced for comparisons, like Jungfraujoch, GAW etc. , have they been influenced by local source or not? You can't conclude that "because of this, the site is likely influenced by local. . ." what "local" is it? 4) The definition of the "background" is far too subjective, what background is it, the troposphere, at which altitude, at what season, the regional or global or just around this site? Because it is above "background", then indicates from "local", I am not with your discussions here.

4. About the diurnal variation The conclusion in page 21954: "The strong diurnal rBC variation shown in Fig. 4 further suggests that the QHL basin is heavily influenced by local rBC sources." First of all, I would suggest to plot the median and percentiles as well in these diurnal figures, especially for the BC mass. From these diurnal plots, I can't see any apparent "local" influence on BC mass, you have claimed that(and it is even not true) there was only a very slight peak around 8am, but based on this you conclude "heavily" influenced by local? In the abstract "The high rBC values and their diurnal behaviour strongly suggest that the QHL area was heavily influenced by local rBC sources." There are no apparent 8am rBC peak in Fig. 4, it may be the simple truth that the "background " level BC mass has been diluted by the boundary layer rise in the daytime but was trapped due to the largely decreased temperature in the night. There are no indications of "local" influence from this simple analysis. How could the "heavily" and "strongly" in the abstract come? Moreover, even if there is no data available for source attributions, you should at least point out or estimate where the pollution may come from, by combining with the local wind and back trajectory analysis, not just simply claim that "local residential activities". Once again, there is no point to compare the diurnal trend at this site with the urban study, how could the traffic activities at this plateau site be similar to that in megacity?

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5. I have no idea what is the reason to plot Fig. 6. How could the BC mass loading be necessarily related to mixing state? This figure is kind of misleading at some point. The mixing state of BC could be largely modulated by the meteorological conditions at this remote site, the mass loading is not the main driver. And the mass loading itself cannot indicate the sources at all, so the conclusions on Page 21956: "This large variability is mainly attributable to the local rBC sources" "drop off of diesel traffic in the evenings, combined with the continued contributions of biomass burning for cooking and heating" are not convincible at all, and even you have not convinced readers what the exact BC sources are.

6. About the BC/CO ratio. The ratio BC/ΔCO should be normally used rather than the BC/CO, because we need to get rid of the fairly large background of CO (which is around 150ppb in this study). This should also apply to the comparison, please check the other literatures carefully, making sure you are comparing the same thing.

The discussions in page 21958-21959 are a bit superficial: an rBC/CO ratio as derived from the whole dataset, comparing with the other literatures, then claim just because of this combined sources are indicated, I think further analysis is essential to make the conclusion solid: 1) how the BC/ΔCO ratio varies with different air masses 2) how could this ratio be influenced by meteorological conditions including the precipitations? 3) how could the BC/CO related to the ageing time of BC, i.e. the mixing state of BC, as the lifetime of BC is mostly shorter than CO, the BC/CO ratio could be modified during transport.

A few more points should be addressed:

1. At least the backtrajectory data analysis needs to be done to get the idea where the BC is from. We need to investigate how the BC mass, size and mixing state, BC/CO vary with different air mass origins.

2. The analysis on source attribution should be robust, not just simply "guess" what are the possible sources. The local wind data along with the backtrajectory data should

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be combined to investigate if the pollutants are local or regional. The current analysis cannot pronouncedly conclude the spatial scale of the pollutants.

3. The final conclusion should be upgraded to be a scientific level, to state why these dataset is important rather than just listing the results.

4. After all of the above points are addressed, the abstract of this paper needs to be revised carefully.

Technical: Page 21950 line 11-15: please state clearly what is the altitude (above sea level) of the experimental site Page 21951: Line 12: could you explain how you get the 25Line 15 and 16: please rewording this sentence Page 21954 line 18: “in contrast”? how could it be in contrast? The peak all occurs around 8.

I am not commenting too much on the technical issues/typos, given many places of this manuscript needs to be rewritten.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 21947, 2012.