

Interactive comment on “Reevaluating the black carbon in the Himalayan and Tibetan Plateau: concentration and deposition” by Chaoliu Li et al.

Anonymous Referee #2

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The authors address an important and underappreciated issue: the effect of mineral dust, more precisely inorganic carbonatic carbon, on black and elemental carbon measurements. The authors present some data on this, but the issue is in the current state of the manuscript not discussed thoroughly enough. Secondly, the authors discuss the role of catchment area influx of BC to lake sediment records, causing potentially flawed inferred atmospheric BC deposition results for the studied area. This second hypothesis of the manuscript is poorly justified and unnecessary for the manuscript.

In more detail, major concerns are:

- The study discusses two quite separate issues, which should be clarified notably in the introduction: First, it is discussed that (apparently mostly) atmospheric measurements of BC (more specifically EC) may be overestimated for the HTP due to mineral

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dust interfering the measurements. Secondly, a quite unrelated issue of lake sediment records being affected by riverine influx of BC and not only atmospheric BC deposition. These both factors may result in over-estimations of BC or EC concentrations in these records. These two study premises should be clearly pointed out. While the hypothesis on atmospheric measurements may be justified, the authors show rash and quite poorly grounded critique on the HTP lake sediments as records of atmospheric BC deposition, as discussed below.

- The language in general is not of the required high quality (particularly in the beginning of the manuscript), and in some places even poor leading to possible misunderstandings. The word “BC deposit” is used incorrectly as a noun (e.g. line 61). It is BC causing the climate impacts, not the environmental record or matrix (i.e. “deposit”, as the authors incorrectly use the term) itself.

- The statements on lines 71-74: “However, the above studies present limitations because of unique environments found in the HTP (e.g., high mineral dust (MD) concentrations in aerosols and catchment inputs to lake sediment). Therefore, the above studies should be re-investigated to better define the actual BC values.” are key to the manuscript but very poorly justified by the authors. For instance, the authors cite Kaspari et al., 2011 as being one of the studies that has limitations due to e.g. high mineral dust concentrations in the HTP. However, Kaspari et al. specifically use a BC quantification method (SP2, i.e. single-particle soot photometer) which is specifically NOT influenced by dust. Furthermore, Kaspari et al. (2011) measured mineral dust separately in the same study by using iron as a proxy for mineral dust. Therefore, these statements by the authors are unjustified, and questions also the other citations by the authors. Significantly higher accuracy is required by the authors before making such bold statements.

- The authors are correct in stating (lines 78-85) that inorganic carbon (IC) may influence the total carbon (TC), particularly organic carbon (OC), and even elemental carbon (EC, thermal optical proxy for BC) concentrations. However, this applies ONLY

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for OC/EC measurements with the thermal optical transmittance/reflectance (TOT/R) method. Furthermore, it depends on the used protocol for the measurements. Cavalli et al. (2010) have studied this issue thoroughly: "Natural calcite evolves in the He-mode with the EUSAAR_2 and the NIOSH protocol, and will be detected as OC, whereas it evolves in the He/O₂-mode with the IMPROVE protocol and will possibly be detected as EC. Neither the NIOSH protocol nor the IMPROVE protocol address definitely this issue of CC. The NIOSH method 5040 recommends fumigation of the aerosol samples with HCl prior to thermal-optical analysis to eliminate any contribution of CC to OC and/or EC signal. However, fumigation with HCl has been shown to cause artificial loss of volatile organic acids (Chow et al., 1993) and to induce intense charring phenomena in ambient aerosol samples (Jankowski et al., 2008). The authors have not addressed these issues appropriately in the beginning of the manuscript. These issues are discussed at the very end of the manuscript which is too late, as readers familiar with these issues may have stopped reading the manuscript after this insufficient introduction.

- One of the main hypotheses of the manuscript is presented on lines 139-155. The authors claim that modelled BC deposition and observed BC deposition in e.g. lake sediments should be of comparable size. As the observed BC deposition in lake sediments is ca. 10 times higher than the modelled value, the authors claim that the discrepancy should be caused by the lake sediments not representing only BC influx from the atmosphere but also from the catchment area. This is the second main premise of the manuscript, but is unfortunately totally unjustified and false. The statement of the authors that the modelled and observed values should at least be comparable, is false. Previous studies have shown several times models to underestimate observed BC concentration and deposition values (2-5 times or even more than by a magnitude) e.g. in the Arctic and China (e.g. Koch et al., 2009; 2011; Bond et al., 2013 and references therein). These under-predictions by models often relate to difficulties in parameterizing, for instance, BC properties, aging, transportation and scavenging efficiencies properly. Moreover, the authors make a major error: observational data

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is data that is used to validate modeling results that are based on estimations of BC emission strengths, and not the other way around. Modeling data is validated to be reliable based on observations, and the models are tuned accordingly. Furthermore, the authors make bold presumptions without any scientific evidence of the higher observed BC fluxes in sediment being caused by catchment influx of BC to the sediment cores. As a reviewer, I strongly suggest that the authors should consider getting familiar with basics of paleolimnology before making bold accusations about lake sediments as records of BC deposition, e.g. "Paleolimnology" by Andrew S. Cohen (2003, Oxford University Press). Basically, there are lakes that are well suitable as BC repositories as they mainly collect atmospherically deposited material. Crucial is, where the sediment core is collected, at the deepest point of the lake and preferably from a lake with very smooth bathymetry. In such a case the sediment core is not at all affected by redistributed sediment e.g. from the catchment area. So the coring site selection is crucial. Additionally, the lake sediments are dated based on lead²¹⁰ deposition. As the amount of lead 210 is known for the present, and its half-life time is known, lead²¹⁰ measurements vertically in a sediment core present a robust dating technique. Subsequently, the amount of lead²¹⁰ in the sediment samples will also inform the researcher whether the coring site is affected by sediment redistribution or not. If not, then the amount of lead²¹⁰ is the same in the surface sediment sample as in the atmosphere. If there is less or more, this is an indication of the coring spot losing or gaining additional sediment and thereby also BC (e.g. Blais & Kalff, 1995). This can be studied by the sediment focusing factor of the sediment core (Blais & Kalff, 1995). So have the authors checked what the sediment focusing factor of the studied HTP lakes is? According to their suggestion of the lakes recording 30 times too high BC deposition flux values compared to the modelled values due to sediment influx from the catchment area would mean that the sediment cores would have to receive 30 times more sediment influx from the catchment area than what they accumulate naturally. Such values are unheard of in these types of research. Consequently, the authors need to get a basic understanding of paleolimnology and lake sediments as records of contam-

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inants before making any such suggestions. Basically, this unfounded premise greatly undermines the half of the manuscript discussing this issue.

- On lines 156-168 the authors present that other studies made based on ice core records show lower BC concentrations and deposition values than the lake sediment and that ice cores are more suitable than lake sediments to record only atmospherically deposited BC. Yes, ice cores record more reliably only atmospherically deposited BC, but when taking into account the sediment focusing factor, input of BC to the sediment cores from the catchment area can be eliminated and values more reliably presenting atmospheric deposition can be achieved (Blais & Kalff, 1995). Secondly, the authors make a mistake in comparing directly lake sediment and ice core BC records. This is because BC has been mostly analyzed with different analytical methods from these archives resulting in different types of BC particles being quantified (e.g. Hammes et al., 2007). Subsequently, even from same samples considerably different BC concentrations can be detected (e.g. Watson et al., 2005). Sure, many HTP ice core and lake sediment records measure BC as elemental carbon with thermal-optical methods from the samples so that the values should basically be comparable. However, the thermal-optical measurements of the sediments undergo extensive chemical pretreatment before the actual BC analysis and this may partly result in different BC particles being quantified. The comparison is not as straight-forward as the authors try to convey. Please, also remember to consider the different protocols in the TO-measurements as the authors have pointed out themselves. This issue is, again, discussed at the very end of the manuscript, which is not a proper structure for this.

- "Because glaciers are generally located at the highest altitudes of a given region, they only receive wet and dry depositions of BC from the atmosphere." This is a very confusing statement (line 161-162) that needs clarification. Low-elevation glaciers can be found around the world. The key is to collect the ice core from the accumulation area of the glacier, not e.g. ablation zone.

- I disagree with the comment by the authors (lines 165-168) that "In addition, because

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the HTP is situated in a remote region, BC deposition patterns in the HTP must be compared to those of other areas (e.g., the Arctic, Europe and eastern China) to better understand the patterns." Europe and especially the Arctic have different sources and deposition processes for BC and comparison of HTP BC deposition with Arctic BC deposition seems unjustified.

- Line 204-205, I hope blank values were subtracted from reported values and not the other way around as suggested in the text.

- The tests of the authors to remove carbonates from the atmospheric sample filters by fumigation are valuable and interesting. However, most useful results could have been achieved if these measurements were conducted using all the appropriate different temperature programs available (e.g. NIOSH, IMPROVE and EUSAAR_2), because all these have been used in literature, and carbonatic carbon evolves at different analysis stages during these protocols.

- The discussion on lines 301-310 indicates that the authors aren't really sure what happens to carbonates after the acid fumigation. Does carbonate affect the OC or the EC quantification? Seemingly both, and varyingly from sample to sample. To me, this causes high uncertainties in the interpretations of the data. This procedure: "samples with BCA/BCO above one was set as one in calculation of the average value at two stations" sounds quite artificial and there is a lot of room for intense charring being caused by the fumigation (Jankowski et al., 2008) obstructing the kind of analysis the authors are attempting.

- The statement on lines 323-324 "In general, the BC deposition levels measured via different methods should be consistent for a given region." is false for even within one environmental matrix, e.g. sediments (Watson et al., 2005; Hammes et al., 2007; Han et al., 2011) or snow samples (e.g. Lim et al., 2014) let alone between different environmental records (e.g. Rose & Ruppel, 2015). It is absolutely inappropriate to compare BC deposition values measured using different analytical methods as these

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measure different BC particle types, as the previously cited work clearly reveals. By comparing suitable selected work with one another on lines 324-336 the authors were able to compare some similar values recorded with different methods for some regions, but at the same time neglected a huge amount of data available which did not suit this statement.

- Section "3.2.1. Overestimated BC deposition from lake cores of the HTP" contains a lot more promising discussion that expected on the false presumptions presented in the introduction. This section could be clarified and made more convincing with inclusion of the sediment focusing factor calculation and discussion from the respective lakes. However, it is unclear why the authors want to compare in Table 2 BC deposition results from all around the world to HTP values? It's inappropriate and it seems that the authors have simply selected sites that support their points and are not presenting all available data. For instance, all Greenland ice core data is neglected, although there is a lot of data available, but it just happens to show almost a magnitude lower BC deposition (e.g. McConnell 2010) than other Arctic results, for which reason it seems not to have been presented here. I strongly suggest leaving away the discussion and data of other areas than HTP and Asia. Particular attention should be paid to the fact with how different methods the results in the different articles are produced.

- Also in Section 3.2.2., please remove any comparison of HTP values to e.g. Arctic or European BC deposition values. Such comparisons are inappropriate due to different methodologies and very different sources and BC deposition processes in these areas. Furthermore, these comparisons are not necessary for this manuscript and don't lift its significance in any way.

- Surprisingly then, the last paragraph of 3.2.2. discusses the possible uncertainties in comparing the BC deposition results between different methods and environmental archives. Good! Unfortunately, this discussion comes too late in the manuscript and is not thorough enough. Things mentioned earlier in this review should be carefully considered.

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All in all, the manuscript contains some important perspectives and a little new data on carbonatic carbon in atmospheric BC samples, but it is questionable whether this is enough data to constitute sufficient scientific novelty for a publication. The amount of carbonatic carbon is measured in some atmospheric samples, but it is not estimated whether or how this would have affected e.g. previous snow and ice core measurements in the HTP, where dust is frequently present. This discussion is majorly hampered by the fact that dust, and therefore carbonate, concentrations vary substantially from sample to the next and will have variable influence accordingly. Much more discussion on these issues is required for the manuscript and even after that, it's maybe enough to publish only as a technical note.

The discussion on BC deposition values reported for the HTP in lake sediments not reliably representing atmospheric BC deposition, and that the BC concentrations and deposition should be re-evaluated in the HTP, is unjustified and poorly researched. The fact that two lake sediment records show different deposition values than for instance ice cores and atmospheric observations is ultimately not surprising. If the authors wish to further study this issue they should first try to deeply understand the factors controlling BC deposition in lake sediments and the importance of different analytical methodologies for the interpretations. All discussion on other than HTP, or Asian, BC deposition results should be removed from the manuscript. In my personal opinion, this lake sediment discussion should be completely removed from the manuscript. It is unrelated to the carbonate issue, and there is so much other, e.g. ice core, BC deposition data available from HTP, that it is unnecessary to try to re-evaluate the whole HTP BC deposition patterns based on these two sediment records showing different results than other records in the larger area.

Consequently, the authors present too little own new data and their discussion on literature data is mostly inadequate and not presented in a clear structure in the manuscript.

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