

Author responses (and description of revisions) to review comments

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

Dear reviewer:

We are grateful for your constructive suggestions and questions, which greatly improve this MS. Despite some critical comments from both reviewers, you give us the overall positive assessments. You provided not only detailed overview but also lots of detailed and important questions and suggestions to the MS. Meanwhile, we also modified some mistakes that found by ourselves during the modification. In addition, the English of the MS has been improved by the professional English editor of Springer nature (Receipt code: GOTRE-F49-0710225832). According to suggestion of English editor, the title of the MS was modified to “*Reevaluating black carbon in the Himalayas and the Tibetan Plateau: concentrations and deposition*” and all the English of the MS was greatly improved.

We show our great thanks to all the questions and suggestions and have answered all of them. Our answers and modifications in the revised MS were marked in blue. The sentence added in the revised MS was marked in red and italic.

Because new experiment has been conducted by help of other researchers from Shandong University, three more researchers were invited as co-authors of this MS.

If you have more questions or suggestions please let us know.

Best wishes !

Chaoliu Li on behalf of all the co-authors

2017/7/20

Point-by-point response to reviewer’s comments

Anonymous Referee #1

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Comments on “Reevaluating the black carbon in the Himalayan and Tibetan Plateau: concentration and deposition” by Chaoliu Li et al :This paper describes a reevaluation of the black carbon content mostly at two sites in the Himalayan and the Tibetan plateau, the Everest and Nam Co stations. The authors consider that the BC concentrations were overestimated due to an underestimation of the carbonates from mineral dust. They also compare various BC measurement methods (in snow and glacier, in lakes and in the atmosphere) to test the coherence of deduced BC concentrations. They found that the BC concentrations measured in lake cores are mainly driven by river sediments and not atmospheric deposition. The adjusted BC concentration is then estimated as ranging from 10-25 mg m⁻² a⁻¹.

Thanks a lot for the detailed overview of the main points of the MS.

Main comments: The introduction is very long and could be better structured to obtain a clear view of the already published results, the problems related to the estimation of BC concentration and the analysis presented in this study.

Answer: Thanks a lot for the suggestion. The other reviewer also show the similar

feeling of the introduction. The introduction has been cut to short and those not very related the topic of the MS were deleted in the revised MS.

- Several species containing carbon are described (IC, CA, BC, OC, EC, TCA: : :) and one subject of this study concerns the wrong estimate of BC. BC is however not the only carbonaceous compound that absorbs -and therefore contributes to atmospheric warming- and that modify the surface albedo – and therefore modifies the glacier persistence. Can you please describe the effect of these various carbonaceous compounds on the climate ?

Answer: Thanks a lot for this question on the different climate forcing effects of different components of the carbonaceous matter. These expressions were double checked in the MS. It need to point out that “EC” is the common chemical/mass definition of “BC”. CA contains BC and OC. Therefore, we added the climate forcing of OC because light absorption of BC has been shown in the MS. Climate forcing of IC discussed in the following question. Meanwhile, replaced “TC” by “CA” in the revised MS and related figures. The sentence “*Organic carbon (OC) is generally considered to scatter sunlight. However, some components of OC also absorb sunlight and warm the atmosphere (Andreae and Gelencser, 2006). Therefore, the contributions of IC to the OC and BC values in the HTP aerosols must be quantitatively evaluated.*” was added into the revised MS.

- For example, you mention (§3.1.2) that the presence of carbonate led to an overestimation of the HTP TC levels. TC= total carbon. Is IC not comprised into “total carbon” ?

Answer: Thanks a lot for the question and sorry of confused expression. “total carbon” contains IC and we changed the description in the revised MS. To make it clear, that sentence was modified to “*IC can also emit CO₂ in response to increasing temperature during measurements, thus causing an overestimation of the total carbon (TC) in carbonaceous aerosols (CAs) (Karanasiou et al., 2010)*”.

Is the effect of IC (once it is converted in equivalent BC) is air different than BC regarding the light absorption and warming of the air? Is the effect of IC different than the BC one regarding the surface albedo of glacier with deposited dust ? It is really important to have a clear discussion of these various effects to demonstrate that your main results (the BC concentration is partially due to the presence of carbonates) is important for climate analyses.

Answer: Thanks for the question. It is better to compare albedo reduction caused by mineral dust (including IC) and BC because both of them belong to light absorption particles. Previous study has shown that BC had higher light absorption ability than that of mineral dust in atmosphere. For instance, Dust particles are more weakly absorbing per mass (about 0.009 m² g⁻¹ at 550nm for Asian dust) (Clarke et al., 2004). The corresponding value for BC is 5 m² g⁻¹ at 550 nm (Bond and Bergstrom, 2006). Therefore, BC has higher albedo reduction ability than that of mineral dust at the surface of glacier (Qu et al., 2014). A sentence “*Because MD has lower influences on light than BC in the atmosphere (Bond and Bergstrom, 2006; Clarke et al., 2004) and on glacier surfaces (Qu et al., 2014), considering IC as BC will overestimate the BC-driven climate forcing.*” was added into the revised MS.

- It seems (§3.2.1) that the fact that BC in lake core is largely influenced by catchment inputs was already demonstrated in several publications. Please specify clearly what is the new input of your research !

Answer: Thanks a lot for the suggestion. A sentence “*although the influence of sediment focusing on BC deposition in lake cores has been noted in other areas (Blais and Kalff, 1995; Yang, 2015), it has not been pointed out and evaluated in the HTP.*” was added into the revised MS and some not important sentences were deleted.

Does the IC content depend on the mineralogical composition of MD? If yes, what are the difference between various minerals? Does it change between the various deserts around the world ?

Answer: Yes, IC content of different minerals are different, because IC is mainly contained in carbonate minerals. Spatially, the dryer the desert, the highest concentration of IC. But for the HTP, the mineral dust is transported mainly from the local sources of itself (Li et al., 2012). Because aerosols are well mixed when transported in atmosphere (Sun et al., 2007), the IC concentration of collected atmospheric particles should be varies little.

- Lines300-305: at this place you mention for the first time that the acid-treated samples transfer OC to BC components. Depending of the rate of this process (that perhaps also depends on temperature), the discussion of figure 2 and §3.1.1 has to be changed. Moreover, this information is really necessary to be explained under §3.1.1

Answer: Thanks a lot for the suggestion, and the other reviewer has pointed out the similar suggestion. The uncertainty of this method has been moved ahead and related explanations have been modified in the revised MS.

- English should really be improved!

Answer: The whole MS has been improved by the professional English editor of Springer nature (Receipt code: GOTRE-F49-0710225832).

Minor comments: - Line 45: please check the language

Answer: The sentence was changed to “*Correspondingly, BC deposition derived from snowpits and ice cores agreed well with those derived from models, implying that the BC depositions of these two methods reflect the actual values in the HTP.*”.

- Lines 71-73:

please rephrase + are you sure that all these studies have methodological limitations bounded to the mineral dust underestimation and the impact of catchment inputs? –

Answer: Thanks a lot for the question, which was also pointed out by the other reviewer. Sorry that there are inconsistencies between two sentences and incorrect references adopted between lines 67-73. Therefore, all of those references not related to aerosol of the HTP were deleted. To make the total expression consistent, the sentence was modified to “*numerous studies have been conducted on the BC concentrations in the atmosphere (Cong et al., 2015; Marinoni et al., 2010; Ming et al., 2010; Wan et al., 2015; Zhao et al., 2013) and atmospheric BC deposition as determined from lake core sediments (Cong et al., 2013; Han et al., 2015). However, all of these studies exhibit limitations because of certain special environmental factors in the HTP (e.g., high concentrations of mineral dust (MD) in aerosols and catchment inputs to lake core sediment).*” In the revised version.

Line98-99: there is probably other species which concentration depends on particle size. Please mention them (see also further comment on BC content as a function of size distribution).

Answer: Yes, other species such as carbonates and related major elements (e.g., Ca, Fe and Mg) are closely connected to MD and particle size distribution. Because we mainly discuss IC in this MS, that sentence was modified to “*Because TSP samples contain more MD and carbonates than PM_{2.5}, they should have higher concentrations of IC*”.

Lines 118-121: It is obvious that the MD content during dust storm is high !+ Please add the reference for this results

Answer: Thanks for the suggestion. Because this part is not very important in the introduction and the reviewer suggest that we cut the introduction. This part was cut in the revised MS. Therefore, the related reference was not added.

-line 142: : : : than the values measured in lake cores at Nam Co and Qinghai lakes: : :

Answer: Suggestions accepted and that part was modified in the revised MS.

- Line 143-144: Do you mean that the HTP BC content is “measured” outside the plateau or that BC content comes from other regions that the HPT plateau ?

Answer: Thanks for the question. The cut the introduction, this part was deleted from the revised MS.

- Line 156: which kind of values? Other measurements? other kind of data? Other analysis and methodologies ?

Answer: Here reliable values is BC deposition value, which was added into the revised MS.

- Line 159-160: which quantity is then measured if they did not report BC deposition directly ?

Answer: Sorry for the confused expression. The mean here is BC deposition can be “calculated out” not “measured”. Therefore, this sentence was modified to “*in those articles*” was added to the end of that sentence.

What do you mean by “deposition pattern” ?

Answer: Thanks a lot for the question. “patterns” is an extra word and need to be deleted. In addition, because the other reviewer thought that part of comparison between the HTP and other regions was useless, that part was totally deleted from the revised MS.

- 172: please delete (Nam Co station)

Answer: “Nam Co station” deleted.

- 204: I suppose the blank concentrations were subtracted from the measured concentrations?

Answer: Yes, your assumption is what described. Therefore, a sentence “All the reported values in this study were corrected based on the values of the blanks.” was added into the revised MS.

- 213: “previously reported BC deposition data were adopted”: what is the meaning of this sentence ?

Answer: Sorry for the confused description. This sentence was modified to “*To determine the actual BC deposition in the HTP, previously reported data were compiled and evaluated (Table 1)*”.

- 225-226: CBC-TSP is then the equivalent BC (usually reported as eBC) concentration

in the atmosphere ?, From where do you take the dry deposition velocity and the particle washout?

Answer: Thanks a lot for the question. C_{BC-TSP} is the abbreviation of BC concentration of the total suspended particle (TSP) for a given region, not equivalent BC. The values of dry deposition velocity and the particle washout were taken from reference of (Fang et al., 2015), which was added in the revised MS.

- 231: the values used in the BC deposition calculations for these two areas are shown: : :

Answer: This sentence was modified according to suggestion.

- 236: this is probably your main results. You have first to present the measurements and then to give the main conclusion. At this place, I can only say that up to now you have not given any proof for this statement.

Answer: Sorry of not good expression. A sentence “*after compared with BC and OC concentrations of original and acid treated TSP samples,*” was added in front of results.

- 239-246: I do not really agree with your statement “BC concentrations are more heavily influenced than OC and TC”: the ratio BC_a/BC_o is lower than for the one of OC or TC, but the uncertainties are really high (0.37 and 0.26) leading to a much more nuanced conclusion! Is it possible that the uncertainty differences between Nam Co and Everest is bounded to another kind of carbonate dissociated at various temperatures ?

Answer: Thanks for the suggestion. This sentence was deleted from the revised MS. We think the uncertainties were because of the different mineral dust concentrations in different aerosol samples at two stations, which was added to the revised MS.

In addition, the other reviewer think those samples with BC_a/BC_o higher than 1 cannot be simply considered as 1. Therefore, the exact ratio were recalculated used the new database.

- Figure 2: since the acid treatment is supposed to remove the IC, how is it possible to obtain values greater than 1 ? It should be really informative to add error bars for both TC and BC ratios.

Answer: Thanks a lot for the suggestion. The other reviewer also pointed out this question. The main reason of the ratios higher than 1 at both stations is that sometimes the treatment of ambient samples by acid make some components of OC easily be pyrolyzed and transferred to BC (Jankowski et al., 2008). However, so far, no perfect method can deal with this problem. Therefore, those samples with ratios above 1 were deleted and the final results were recalculated. Meanwhile, a sentence “*Nevertheless, the ratio of BC_a/BC_o was considered to be slightly overestimated as some portion of OC was considered BC in the acid-treated samples (Jankowski et al., 2008)*” was added in the revised MS to point out the uncertainties. Because each point of Fig. 2 represented only one sample, so that no error bar was shown on the figure. Furthermore, to make the figure more clear. The ratios for TC and BC during monsoon period and non-monsoon period were added on Fig. 2.

- 254: I suppose that you consider only MD and TCA as components of your sample, leading to $MD+TCA=100\%$. The mention of percentage in the text (and not only in the figure) will clarify this point. Could you also give the uncertainty on dust and TCA percentages in order to estimate if the difference between non-monsoon and monsoon seasons are significant or not ?

Answer: Thanks a lot for the suggestion. “TCA” should be “CA” in the revised MS. The samples contain other components other than MD and CA. Here we study only the relative ratio of MD and CA. To make it clear, a sentence “*To evaluate the relative ratio of MD and CA, MD/(MD+ CA) values were calculated (Fig. 3)*” was added in the revised MS. Difference of MD/(MD+ CA) between non-monsoon and monsoon seasons of samples of two stations were checked. The results showed that the difference at Nam Co station was significant ($p < 0.01$), but at Everest station was insignificant ($p > 0.05$), which was also added into the revised MS.

- 258: it seems to me that the NCO-P station was not mentioned before, is not in Figure 1 or in Table 2 ?

Answer: Sorry that location of NCO-P station is not mentioned in both of them. Therefore, location of this station (27.95°N, 86.82°E, 5079 m a.s.l) was added in the revised article when it first appeared.

- 262: does it mean that the NCO-P station measures PM10 and not TSP ? Was the method applied at NCO-P the same as at Nam Co and Everest station ?

Answer: Yes, that research only reported data of PM₁₀ and PM₁. Therefore, data of PM₁₀ was adopted for the comparison with this study.

The methods used in that study was different from this MS. For instance, Ca and Mg concentrations of that study were only water-insoluble fraction, which will cause underestimate of mineral dust (Decesari et al., 2010). Meanwhile, EUSAAR_2 temperature program of Thermo-Optical Transmission (TOT) method was used in that study for OC and EC measurement, which was also different from method in this article. Therefore, a sentence “*because the measured particle size (PM₁₀) and the measurement methods of Ca, Mg and EC at the NCO-P station differed from those in this study (Decesari et al., 2010), uncertainties exist in such a direct comparison*” was added into the revised MS.

- 265: do you have a reference estimating the contribution of local surface soil to total MD contribution? How is it possible to distinguish between local and desert contribution to MC ? by the carbonate types ?

Answer: Yes, according a previous study at this station, even those fine particle in the atmosphere was derived mainly from local source soil (Liu et al., 2017). This reference has been added into the revised MS. Rare earth element ratios can be potential indexes to distinguish local and desert sources. For instance, in our previous research we found MD of the Tibetan soil itself is the main sources of particles loaded at glacier of the Tibetan Plateau by these indexes (Li et al., 2012). Sorry that we do not find related knowledge on how to use carbonate types to distinguish MD sources.

-267-268: please rephrase - 269-272: you compare precipitations at Nam Co and Everest during one year (January 2014-January 2015), that do not correspond to the unique monsoon period. I suppose that most of the precipitation occurs during monsoon, but this should be specified.

Answer: Thanks a lot for the suggestion. The precipitation of two station were modified to that of monsoon period in the revised MS. Accordingly, the sentence was modified to “*the precipitation level recorded at the Everest station (172 mm during the monsoon period between 2014 and 2015) is much lower than that of the Nam Co station (258*

mm), causing high MD concentrations in the atmosphere of the Everest station during that period”.

- 275: you mentioned under §2 that you measured soil samples. The PH is however taken from a reference. Were the same samples used for both studies?

Answer: Not the same samples for two stations. Data of one study at Nam Co region is taken from our previous article (Li et al., 2008), therefore the reference was adopted. Correspondingly, pH of soil sample collected around Everest station was newly measured, which was mentioned in method part (last paragraph of the 2.1).

- 279-282 + Figure 4 : The slope of Ca versus IC is smaller (about 0.25) for Nam Co than for Everest station (0.30). Does it mean that there is different carbonates types at both stations ?

Answer: Yes, your assumption is correct and the corresponding explanation was added into the revised MS. Therefore, a sentence *“The ratio of Ca/IC was higher in the Everest station samples than that of Nam Co station, possibly reflecting different types of carbonate at these two stations.”* was added into the revised MS.

- 289-292: here again, it is not possible to ignore the very high uncertainties in the conclusion of the impact of IC on BC concentration.

Answer: The exist of uncertainties for this conclusion was added into the revised MS. Therefore, a sentence *“Moreover, because of the large variations in the above values, the corrected BC concentrations at the two stations have large uncertainties.”* was added into the revised MS.

- 294-292: was the BC concentrations measured by Ming and Cong estimated with a similar method that your method described in this paper ?

Answer: Yes, this MS used the same method (Thermal-Optical method, IMPROVE-A) with those of two studies, which was added in the revised MS.

- 294-292: please report the uncertainties of BCa/BCo to the estimated concentration in ng/m³.

Answer: Thanks for the question. The RSD of BC concentrations of the two stations were shown based on that of BCa/BCo.

- 296: does the decomposition temperature of carbonate depends on the size of the particle ? If yes, you have to put a reference.

Answer: Related studies were double checked and did not find the direct proof. Therefore, this sentence was deleted in the revised MS. Nevertheless, previous study showed that IC of carbonates could influence both OC and BC of aerosols (Karanasiou et al., 2015).

Therefore, this sentence was modified to *“The OC concentrations in the treated samples used in this study also decreased, indicating that carbonates can also decompose at low temperatures (Karanasiou et al., 2010)”*.

- 296-300: you mixed several notion if this sentence: please clarify if you want to link the particle size distribution with the decomposition temperature, the BC and OC overestimation with the size distribution or the BC and OC overestimation with the temperature!

Answer: Thanks for the suggestion. This sentence has been rewritten and the part on fine particle size was deleted. The new sentence has been shown in the answer to the

above question.

- 308-310: it seems obvious that carbonate contribute to TC since it was stated before that it contributes to BC.

Answer: Yes, it is just repeat the issues have been pointed out in the MS. Therefore, this sentence was deleted in the revised MS.

- 312: where: not grammatically correct

Answer: Thanks a lot for the suggestion. That sentence was changed to “*Since the influence of carbonate carbon on TC has been observed in PM_{2.5} samples from Qinghai Lake, Northwest China (Zhao et al., 2015), this phenomenon should be clear in the TSP samples in this study.*”.

- 311 -315: could you please mention where are the station of Zhao and Karamasiou so that the reader can estimate why the dust storms are more severe at the studied stations. Please provide the same information for used stations in Cao and Ho

Answer: Study of Zhao et al., (2015) was conducted at Qinghai Lake, West China, where is near to numbers of deserts. Study of Cao et al., (2005) was conducted at city Xi’an, Middle west China, where dust storm happened frequently during Spring. Study of Ho et al., (2011) was conducted at Tongyu, Northeast China and focused mainly on dust storm events. Study of Karamasiou et al., (2015) is a review that discussed the influence of carbonate on OC and BC, so that no station can be pointed out and this reference was deleted here.

Therefore, the locations of the first three stations were added into the revised MS.

- 319: you have to explain why you estimate the overestimation of “at least 45%”

Answer: Sorry of not expressing it clear and making a mistake. As our newly calculated value, the correct expression should be 52%, which was the newly calculated value. Therefore, that sentence was modified and the reason of 52% was added in the revised MS.

- 320-321: please see the main comment concerning the effect of carbonate or BC on climate analyses.

Answer: Thanks a lot for the suggestions. The reply has been shown in the answer to previous question. The relative climate forcing has been added in the revised MS.

- 318-330: if you mention the name of the stations, the reader will take much less time to understand your comparison and to find them in Table 2.

Answer: Thanks a lot for the suggestion and the station names have been added at related positions to make it clearer.

- 331-336: the conclusion is too simple since you have no real correlation between measured BC in the atmosphere and in the lake core and no proof of the polluted east asia and less polluted area in Europe! Moreover §3.2.1 shows that lake core are influence by catchment area and rivers so that you cannot here make some conclusion about Sweden lake core or about deep ocean sediment samples without taking the other results of your paper into account.

Answer: Sorry about the not exact expression and thanks a lot for the suggestions. The other reviewer also suggested that data of Europe and East Asia are not closely related to the HTP. Therefore, we will just use the data of East Asia to prove that BC deposition

of lake sediment of this area can reflect actual atmosphere deposition, but not classify them as “polluted” or “less polluted” areas.

- 340: re much ?

Answer: Sorry about this mistake. It should be “*were much*” and modified in the revised MS.

- 338-344: a usual structure for a paper is not to propose an interpretation at the beginning, but to describe the results and then to draw conclusion! Do you think that you have to take into account some other parameters such as the evaporation of the lake that depends on temperature, surface and depth ?

Answer: Thanks a lot for the suggestion. A sentence “*although the influence of sediment focusing on BC deposition in lake cores has been noted in other areas (Blais and Kalff, 1995; Yang, 2015)*” has been added as the first sentence to change the structure.

- 343: what is NMC09 ?

Answer: NMC09 is a lake core named by the adopted reference (Wang et al., 2011). Therefore, “Fig. 5” was added to the end of the sentence to show its location and reference.

- 347: why the BC concentration in PM_{2.5} should be lower than in TSP? is this statement is clear for MD due to their large size, it is not directly applicable to BC, soot being usually quite small particles. The following conclusion has therefore to be more seriously validated.

Answer: Thanks for the valuable question. It is natural that BC is emitted from combustion activities and large amount of BC exist in PM_{2.5}. However, BC will absorb on the large particle during transportation so that some part of BC will also exist in particles large than PM_{2.5}. For instance, BC concentration of PM₁₀ of urban cities of Helsinki, Finland (Viidanoja et al., 2002) and Lhasa, China (Li et al., 2016) are higher than those of PM_{2.5}. Therefore, this phenomenon must exist in remote area like studied stations. Therefore, a sentence “*Because PM_{2.5} does not include all particles in the atmosphere, the actual BC concentration in the atmosphere should be higher than that of PM_{2.5} (Li et al., 2016; Viidanoja et al., 2002)*” was added into the revised MS.

- 352-355: how can you conclude that the atmosphere and the lake core should have a similar concentration of BC ? This is based on 2 not proved inputs. Moreover 65% «< 100%!

Answer: Sorry about the incorrect expression. “*concentration*” should be “*deposition*” and was modified in the revised MS.

- Figure 5: the axes are not readable.

Answer: The axes of Figure 5 have been modified.

- 407: you probably want to compare BC in ice with BC in atmosphere ? please change the sentence.

Answer: Thanks a lot for the suggestion. This sentence was modified to “*BC deposition rates derived from ice cores and snowpits are proposed to be closer to the actual atmospheric values in the HTP.*”

- 418: the values of BC in remote areas depend on the sources and long range transport processes and cannot be compared without caution.

Answer: Thanks a lot for the suggestion, which the other reviewer also pointed out. Therefore, comparison between BC deposition of the HTP and other remoter areas were deleted from the revised MS. In addition, the last sentence of the paragraph was modified to “*In summary, despite some uncertainty associated with the remote study area, the atmospheric BC deposition rate of $17.9 \pm 5.3 \text{ mg m}^{-2} \text{ a}^{-1}$ in the glacial region of the HTP is proposed.*” in the revised MS.

- 423: it is perhaps better to give the discrepancy in % instead of in mass concentration

Answer: The value was modified to “ *$17.9 \pm 5.3 \text{ mg m}^{-2} \text{ a}^{-1}$* ” in the revised MS.

- 466: if the lake are smaller, the catchment inputs are probably also smaller of the flow through the lake is larger involving a smaller deposition into the lake core. It is therefore not obvious that smaller lake with have a higher BC concentration due to catchment inputs.

Answer: Thanks a lot for the suggestion. To reduce the uncertainty the sentence “The lake cores examined in this study were drilled from the two large Qinghai and Nam Co Lakes, and lakes of this size should only be slightly influenced by catchment inputs. Therefore, the catchment inputs into smaller HTP lakes should be more intense, which should be considered in future studies. ” was deleted from the revised MS.

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