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Title: A newly identified calculation discrepancy of the Sunset semi-continuous carbon analyzer

1. Comments in RC C296:

The authors discovered an error in an older version of the calculation program, which had been designed for the NIOSH method at the time, not meant for the IMPROVE method. Rather than do such an extensive study, it would have been better to simply contact the manufacturer to either alert them, or obtain a current corrected version. In fact, toward the end it appears they did this, yet only presented the results with no comments.

For final publication, the manuscript should be rejected - due to the fact that few users would still have such an older version, it seems of little interest even to the users of such an instrument, and therefore certainly not of general interest.

Our replies:

We appreciate the anonymous referee for the comments and suggestions. However, we could not agree with the reviewer in several aspects, as illustrated below.

Firstly, in our opinion, it was the whole process of identifying, characterizing and solving problems that was scientifically meaningful. Thus, even if the newer version of calculation software has solved this problem perfectly (which is not the case, see Paragraph 4), this manuscript was still not meaningless. On one hand, it explored how the baseline correction procedure of the Sunset instrument worked and how different methods could influence the results. It helped others to know better about the principle of this instrument, which was the premise to optimize instrument settings for any specific situations. On the other hand, it illustrated that the baseline correction procedure, which was usually considered as relatively accurate before, could also cause significant discrepancies with improper methods. The experience summarized here could serve as reference for other instrument validation work as well.

Secondly, the reviewer suggested that the old version of calculation program “had been designed for the NIOSH method” and “not meant for the IMPROVE method”, which was not true. It was unreasonable that the data processing program be specified to every temperature protocol. Thus both older and the current version of calculation program were designed for all temperature protocols, and should be responsible for the results

of all temperature protocols. In fact, it was clearly stated on the homepage of Sunset Laboratory Inc. that the Sunset carbon analyzer is “ready for use with the NIOSH method 5040, IMPROVE and EUSAAR 2 protocols” (<http://www.sunlab.com>). In addition, the calculation discrepancy we reported here was not resulted from the choice of temperature protocols.

Furthermore, the current version of calculation software could not thoroughly solve this calculation discrepancy, even with the NIOSH-like temperature protocol. As shown in Fig. S2, the current calculation program used a piece-wise baseline correction methods. When total carbon concentration determined by multi-point correction method exceeded a determined threshold concentration ($\sim 25 \mu\text{gC}/\text{m}^3$), the baseline correction method changed from multi-point method to a new method similar to single-point method. However, the arbitrary choice of the threshold concentration was the problem. As we have explained in the manuscript, threshold carbon load could vary with temperature protocols as well as sample types. For the two IMPROVE-like protocols the threshold was too large (Fig. S2). For rtNIOSH protocol, we supplemented some experiment of summer ambient samples (Fig. S3). Supplementary sampling was conducted from June 1st to August 31st in 2013, with rtNIOSH protocol and time resolution of 1h (40 min sampling + 20 min analysis). As can be seen from Fig. S3, for rtNIOSH protocol, the threshold carbon load was significantly lower in summer (close to zero) than in winter ($\sim 25 \mu\text{gC}$). Although the current calculation program performed better for winter samples, it failed to identify and correct the calculation discrepancy in summer samples. Thus we recommend that **the occurrence of this calculation discrepancy should always be checked** when this instrument was applied to new temperature protocols or ambient samples with unknown distribution of thermal stability. **This part of discussion was also added in the revised manuscript (See the last paragraph in Section 3.4, Line 374~391).**

In a word, this manuscript was scientifically meaningful in the exploration process of the fundamental instrument principle itself, and in that the current calculation program was not a perfect solution of the calculation discrepancy we identified.

That is all of our responses to the comment from Anonymous Referee #1. We appreciate the time and effort of the referee. Thank you for your kind consideration.

Sincerely yours,

Guangjie Zheng

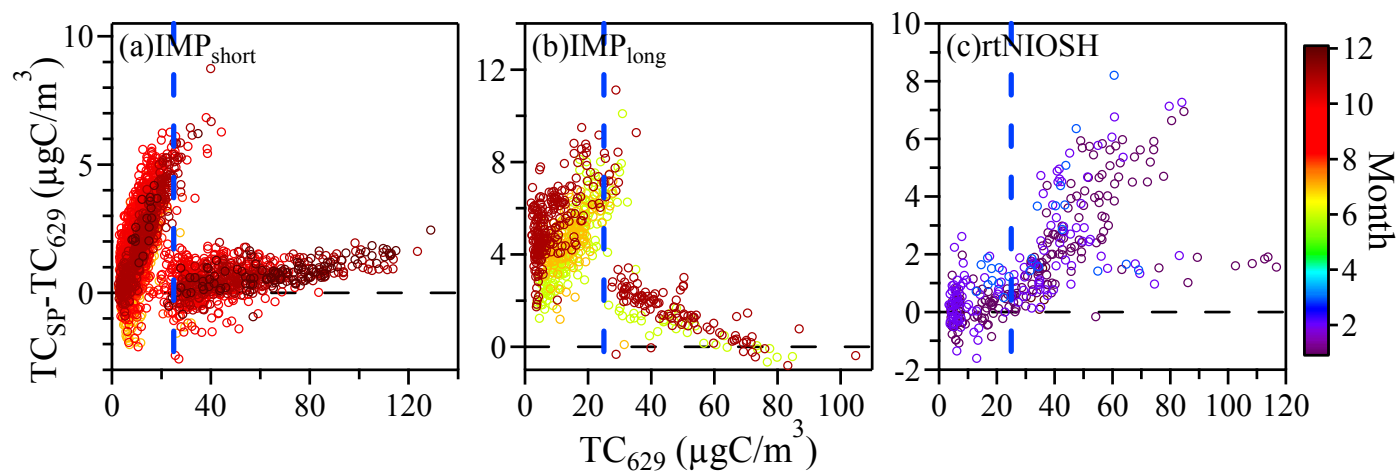


Figure S2. Comparison of TC concentration results given by the newest version (629) of calculation software (TC₆₂₉) and the single-point correction (TC_{SP}), for ambient samples analyzed with (a) IMP_{short}, (b) IMP_{long} and (c) rtNIOSH protocol

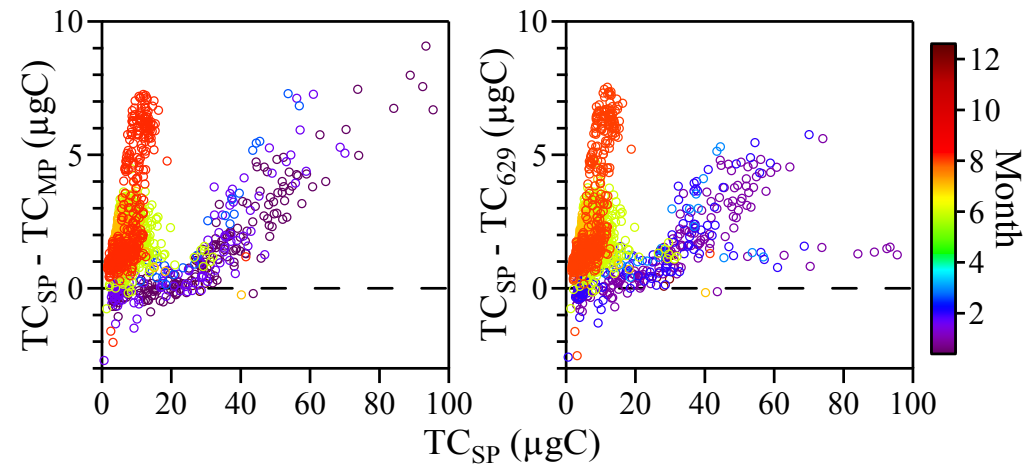


Figure S3. Seasonal variation of threshold carbon load for rtNIOSH protocol. Differences of TC concentration results given by the newest version (629) of calculation software (TC₆₂₉), by the multi-point correction (TC_{MP}) and by the single-point correction (TC_{SP}) were compared.