

Author Comment with regard to:

“Estimation of atmospheric total organic carbon (TOC) – paving the path towards carbon budget closure”

by M. Yang and Z. Fleming

Many thanks for the thoughtful *comments and suggestions from Anonymous Referee #1*. We are very glad to hear that the referee found our contribution valuable, our measurement approach interesting and scalable elsewhere. Below are our replies to the referee’s comments, which are in *italic*.

Anonymous Referee #1

The authors employ a new catalyst based approach to measuring TOC+CO in ambient air (with co-location of high-precision measurements of CO₂ and CH₄ which allow the subtraction of these species). Measurements of TOC are much-needed and though there are limitations to these specific measurements, this manuscript describes an interesting new approach to TOC measurements and a straight-forward application to a marine site.

I have only minor suggestions and requests for clarification:

1. Given the novel measurement approach it seems this should be described in the abstract (brief description, precision, comment on whether all species are comprehensively detected – see comment #2).

Thanks for the suggestions. We will add the following sentences to the abstract (after the first sentence, line 13).

“Here we present a novel and simple approach to measure atmospheric non-methane total organic carbon (TOC) based on catalytic oxidation of organics in bulk air to carbon dioxide. This method shows little sensitivity towards humidity and near 100% oxidation efficiencies for all VOCs tested. We estimate a best-case hourly precision of 8 ppb C during times of low ambient variability in carbon dioxide, methane, and carbon monoxide (CO). As proof of concept of this approach, we show measurements of TOC+CO during August-September 2016 from a coastal city in the southwest United Kingdom.”

We will add this sentence to the second to last line of the abstract:

“Finally, we note that the use of a short, heated sample tube can improve the transmission of organics to the analyzer, while operating our system alternately with and without a particle filter should enable a better separation of semi-volatile and particulate organics from the VOCs within the TOC budget.”

2. Section 2: The authors mention the inlet and briefly allude to SV/IVOCs and aerosols in the text (lines 100, 123-126). The manuscript would benefit from more discussion of this, but most importantly, the authors should reiterate these gaps in the conclusions and abstract. Ultimately the reported TOC is not comprehensive and this should be made clear to the reader, with appropriate suggestions for assessing the degree of

comprehensiveness and/or improving the instrumentation in the future (as given on lines 308-309).

See above. Also, we will add the following sentence to line 308:

“Due to the use of a long, unheated Teflon inlet tube, the semi-volatile and particulate organics were likely not well represented in the measurements presented here. A simple improvement to this measurement system would be to minimize the length of the inlet tube before the catalyst.”

3. Section 2.1: There is some ambiguity of units in this section between ppb and ppbC and it would be useful for the units of all quantities to be carefully defined (line 115). I believe that some quantities are incorrectly given as ppb instead of ppbC (line 131, 135, 141, 145, 148, 151, 155), though it's not always clear from the text. Please correct as necessary.

We apologize for the ambiguity. The units are:

Line 115: ppb C and ppb are equivalent here

Line 131, 135, 141, 145, 151, 155: ppb C

Line 148: ppb C and ppb are equivalent here

These units are specified in the revision.

4. line 84: I suggest you place “e.g” in front of the Nolscher et al. reference since many studies have discussed the “missing OH reactivity”

Suggestion accepted.

5. line 194: September 8-10 also looks windy and rainy from the plots. Why aren't these dates included here?

These dates will be added to this sentence in the revision.

6. Figure captions: I recommend adding the measurement location to the captions so that the casual reader is clear that these represent field measurements at a given site.

Suggestion accepted.

7. Figure 7, Figure 9, and lines 225-226, 250, 263: These scatter plots show some relationships, but the correlation appears quite weak. Please include the R2 on the figures and temper the text accordingly.

The correlations in the scatter plots are heavily influenced by the random noise in the measurement. Take Figure 9, for example, (TOC+CO vs. Sum(VOC)). The r^2 value is 0.12 for the hourly average (N=299), 0.20 for the 6-hr average (N=61), and 0.91 for the bin average shown in Figure 9 (N=6). All of these correlations are highly significant at the 95% confidence level according to a t-test. The purpose of bin-averaging is to remove most of the random noise in order to more clearly demonstrate the mean relationship. We will add this information to the revision.