Response to review #1

Dear Editor, this MS presents a statistical assessment of an alternative method to quantify secondary organic carbon (SOC) in ambient air samples. This method is an alternative to the classic EC tracer method. It is a useful assessment of an alternative method which seems to perform rather well, and therefore merits publication. Reading is somewhat complicated due to the frequent use of abbreviations (eg, fSOC), though. A more fluent writing style would help the reader.

Author's Response: We add a table (also shown below) in the revised main text to help readers to have a quick check of abbreviations used in the paper. We believe this would be more reader-friendly than looking for definitions that scattered in the main text. Please see below for point-by-point response to reviewers' comments.

Table 1. Acronyms and Abbreviations

Abbreviation	Definition
EC	elemental carbon
EC_1, EC_2	EC from source 1 and source 2 in the two-source scenario
f_{EC1}	fraction of EC from source 1 to the total EC
f_{SOC}	ratio of SOC to OC
MRS	minimum R squared method
MRS'	A variant of MRS that use EC from individual sources as input
MT	Mersenne twister pseudorandom number generator
n	sample size in MT data generation
OC	organic carbon
OC/EC	OC to EC ratio
$(OC/EC)_{pri}$	primary OC/EC
$OC/EC_{10\%}$	OC/EC at 10% percentile
OC/EC_{min}	minimum OC/EC
$OC_{non\text{-}comb}$	OC from non-combustion sources
PDF	probability density function of a distribution
POC	primary organic carbon
ROA	ratio of averages
RSD	relative standard deviation
RSD_{EC}	RSD of EC
RSD_{POC}	RSD of POC
RSD_{SOC}	RSD of SOC
SOC	secondary organic carbon
SOC_{svP}	SOC formed from semi-volatile POC
Υ_pri	ratio of the $(OC/EC)_{pri}$ of source 2 to source 1
ε_{EC} , ε_{OC}	measurement uncertainty of EC and OC
Υ_{unc}	relative measurement uncertainty
γ_{RSD}	the ratio between the RSD values of $(OC/EC)_{pri}$ and EC

Some specific comments:

- line 75: I believe Pio et al propose yet another method, using a subset of samples with 5% lowest ratios and discarding the 3 lowest... I don't have the exact reference right now, but please add.

Author's Response: Suggestion taken. The reference (Pio et al., 2011) is now added in the main text. Pio et al. (2011) suggested using the lowest 5% subset to obtain the $(OC/EC)_{pri}$, and if the sample size of 5% subset is less than three, the lowest three data points are used to determine $(OC/EC)_{pri}$.

- line 90: any reason why the Millet method was overlooked?

Author's Response: One reason is that Millet's original paper focused on VOCs, and the MRS approach was used to calculate primary ratio of VOCs/EC to differentiate primary and secondary

VOCs. A second reason we believe is a lack of evaluation work for this method. As a result, the approach initially proposed by Millet et al did not draw much attention from the OC/EC measurement community.

- line 211: please elaborate on why the OCEC10% method provides worse results

Author's Response: Based on the observational data we have, the ambient conditions most likely falls into the scenario between scenario A and B (Figure 3). As such, $OC/EC_{10\%}$ is further away from the true OC/EC_{pri} than OC/EC_{min}, resulting larger bias.

- line 226: I don't understand the different behavior of the $OCEC_{10\%}$ amend the $OCEC_{min}$ methods, given that they are both subsets of the total dataset with specific characteristics of representing 1% and 10%. Why is their behavior different?

Author's Response: Change of f_{SOC} not only changes the position of OC/EC distribution relative to OC/EC_{pri} distribution, but can also alter the width of OC/EC distribution. Because the subset methods rely on percentile of OC/EC, once the OC/EC distribution is widened, the relative position between OC/EC_{min} and $OC/EC_{10\%}$ is also changed and this results in a non-linear response in SOC differences,

- section uncertainty: with some analytical methods (e.g., TOT) the uncertainty is mostly constant (0,1-0,2 micrograms/cm2), please discuss how this would affect the results in this section.

Author's Response: Under the scenario of constant absolute uncertainty, the performance of MRS (Figure R1, 0.2 μg m^{-3}) is similar to that assuming a fix proportional measurement uncertainty (Figure R2, 10% measurement uncertainty). Both Figures R1 and R2 will be included in the revised main text.

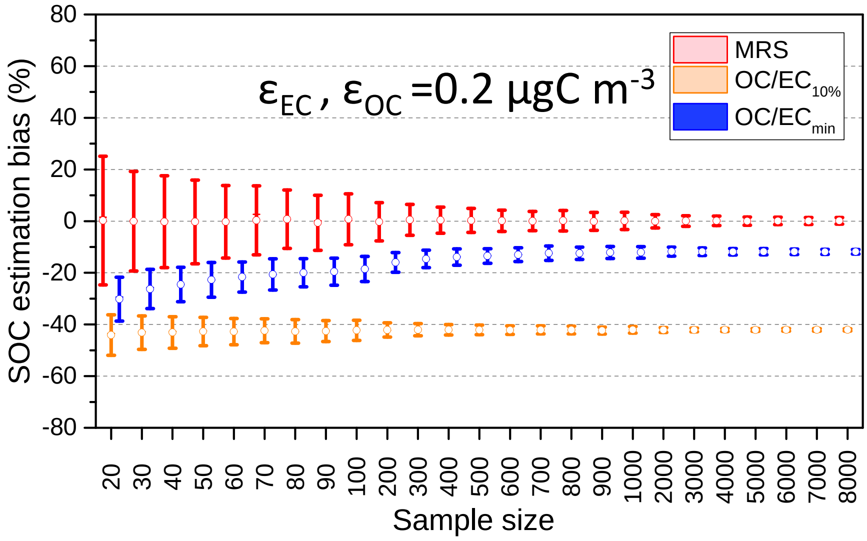

Figure R1. SOC estimation bias as a function of sample size, assuming fixed absolute measurement uncertainty for OC and EC (0.2 μgC m^{-3}). For each sample size, 500 repeat runs were conducted. The circles represent mean of 500 repeat runs, the whiskers represent one standard deviation. Parameters used for testing: Repeat runs = 500; N = 20~8000; EC = 8±4 μgC m^{-3}; $(OC/EC)_{pri}$ = 0.5; POC = 4 ±2 μgC m^{-3}, f_{SOC} =40%, and SOC = 2.67±1.33 μgC m^{-3}.

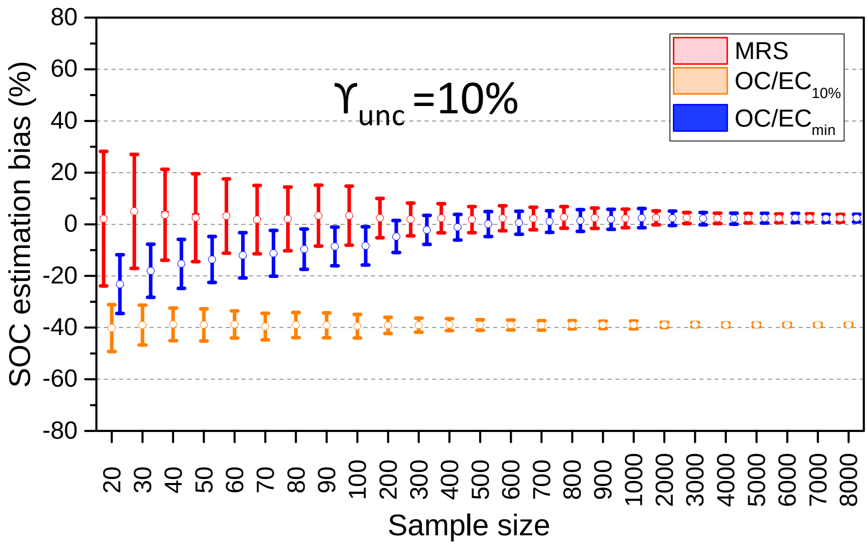

Figure R2. SOC estimation bias as a function of sample size, assuming a fixed relative measurement uncertainty of 10% for OC and EC. For each sample size, 500 repeat runs were conducted. The open circle represents the mean of 500 repeat runs, and the whisker represents one standard deviation. Parameters used for testing: Repeat runs = 500; N = 8000; EC = 8±4 μgC m^{-3}; $(OC/EC)_{pri}$ = 0.5; POC = 4 ±2 μgC m^{-3}, f_{SOC} =40%, and SOC = 2.67±1.33 μgC m^{-3}.

- line 317, please clarify what the authors mean by "the irrelevance of EC and SOC", it is unclear to me

Author's Response: We now rephrased as "the independence of EC and SOC", by which we mean that SOC and EC come from uncorrelated sources.

Reference

Pio, C., Cerqueira, M., Harrison, R. M., Nunes, T., Mirante, F., Alves, C., Oliveira, C., de la Campa, A. S., Artinano, B., and Matos, M.: OC/EC ratio observations in Europe: Re-thinking the approach for apportionment between primary and secondary organic carbon, Atmos Environ, 45, 6121-6132, DOI 10.1016/j.atmosenv.2011.08.045, 2011.