

## Anonymous Referee #1

Received and published: 16 May 2019

Review of “Inter-comparison of the elemental and organic carbon mass measurements from three North American national long-term monitoring networks” by Chan et al. This paper summarizes collocated organic and elemental concentrations from three different types of analysis and sampling protocol. The results are useful in furthering our understanding of thermal optical analyses and resulting biases from sampling artifacts as well as temperature protocols. The paper is fairly well organized and written but could benefit from some clarification. I recommend publication after the authors address comments below.

>> The authors appreciate the useful comments and suggestions from the referee, and we address all the comments accordingly.

Line 1: The title, as well as some description in the text is somewhat misleading because it implies that large geographic scale comparisons are made when in fact the comparisons only exist at one site. Perhaps changing or including something regarding different analytical protocols would help clarify this point.

>> The authors have modified the title to reflect the fact that the inter-comparison was done in one “co-located” site. Corresponding sections, including abstract and introduction have also been modified to reflect this information.

Line 23: Please state years.

>> This coverage information (2005-2015) has been included.

Line 23-26: Again, similar to the title, point out that collocated samplers only exist at one site, so really what is being compared here are the impacts from different sampling and analytical protocols, not a large scale geographic comparison.

>> This is addressed.

Line 29: More on this later, but I don’t understand the value of the normalized comparison. The agreement depends on what you have normalized each time series to. Over what time periods were these comparisons made?

>> In the original study, each time series is normalized to its corresponding concentration measured on Jan 2008. This converts all concentration time series to a percentage change with respect to the measured concentration on Jan 2008. The comparisons for normalized concentration were made from 2008 to 2015.

After some consideration and discussions among the co-authors, we agree to remove the normalized comparison section and focus on the comparison on the absolute data.

Line 35: Is there any evidence for linkages to forest fire emissions and increased vehicular emissions? Did the authors include analysis of these emissions or is this conjecture?

>> Due to the length limit, it is not possible to include all the analyses in this manuscript. A separate analysis that involved the investigation of the 10 years BC emission trends at a number of CABM sites, including a boreal forest site, has suggested that the elevated BC emissions during summer at Egbert could be contributed by the forest fire emissions. The analysis is still currently on going and the full results is expected to be given in a separate manuscript.

Line 36-37: This may be true depending on artifact corrections and how they are applied across a network.

>> The abstract has been modified and this does not apply anymore as the original sentence was removed.

Line 38: Again, extrapolating data and comparisons from one site to “regional to continental-scale-harmonized maps” hasn’t been shown here and may not necessarily be true, especially given different sampling times and sources.

>> The main idea of mentioning a “regional to continental-scale concentration map” is trying to express the effort of evaluating the consistency and compatibility from difference datasets by different networks when using atmospheric OC and EC measurements to constrain their emission changes at regional and continental scales. The “regional to continental-scale concentration map” is not proper and concise expression. It has been removed from the revised version.

The authors have included additional text in the introduction of the revised manuscript to explain the rational accordingly.

Line 23-41: I think it would be helpful if the abstract more closely reflected the comparison work rather than sources which was a rather minor part of the work and mostly based on previously published work (e.g., secondary aerosol formation in summer, smoke in summer, higher OC and EC in summer, etc.).

>> The abstract has been modified.

Line 52: OC can also absorb solar radiation.

>> The word “primarily” is now added.

Line 52,53,56,57: I would suggest using either BC or EC and keeping the same nomen-clature throughout the paper, unless the authors are actually referring to different measurements, then clarify and define.

>> BC and EC share lots of similarity in describing the physical appearance of the aerosol. In some cases, the word BC and EC can be inter-changed but not in all. We do not agree that all the term “BC” in the manuscript can be replaced by “EC” without changing the original meaning. The usage of BC and EC has been addressed in “introduction” of the revised version, and the authors have clearly defined the definition according to Petzold et al (2013).

Line 58: Include “impacts of” changing emissions since OC and EC measurements don’t directly determine emissions.

>> This is addressed.

Line 61: The first sentence is unclear. Wouldn’t long term measurements just depend on making the same measurement over time and doesn’t really depend on a universal definition?

>> In fact, it is challenging to make ambient BC measurements. The word “long-term” is used in this manuscript because the main focus of this work is on long-term measurements. The corresponding sentence has been modified to avoid confusion.

Line 67: The sentence starting with “BC is a generic term” would be a better starting sentence for this paragraph and the authors could remove the current first sentence.

>> This is addressed.

Line 71: Replace “being” with “is”

>> This is addressed.

Line 72: Include “as” after “EC is referred to”

>> This is addressed.

Line 75: Can the authors clarify what they mean by “EC and BC resembled each other”?

>> We meant the trends in long-term time series of EC and BC concentration resembled each other. The sentence is now revised.

Line 80: I am not sure what the authors mean by “direct measurement of carbon mass as part of gravimetric mass”?

>> We meant the carbon mass measured by TOA or TEA is part of the particulate matter mass. This sentence is revised.

Line 97: I’m not sure what is meant by “resulting EC method”?

>> This sentence is revised.

Line 111: Can the authors provide a reference for the OC overestimation?

>> A reference is added.

Line 120: The acronyms for the various networks should be spelled out at first usage.

>> The acronyms of the three networks were first spelled out in the abstract. They are spelled out again in the revised version when it first appear after the abstract.

Line 122: Again, this is somewhat misleading. Add that these collocated measurements occurred at one site.

>> This is addressed.

Line 123: I might have missed this later, but what are the solutions for improving the compatibility?

>> The authors thank the referee for pointing this out and the word “solution” should not be used in here. Instead, the authors have replaced this by “suggestions”. Based on the current work, two suggestions are: (1) ensure maintaining the same SOP for sampling and analytical procedure for any lab to ensure internal consistency, (2) to establish or include the use of a reference material or calibration materials (as suggested by World Meteorological Organization scientific advisory group) during the inter-comparison study. These information has now be included in the revised manuscript.

Line 124: I am not sure the results from one site have been demonstrated to create a regional and continental scale harmonized carbon concentration data set.

>> The authors realized we may not be expressing ourselves clearly and led to misunderstanding. The abstract has now been modified to remove those sentences. The corresponding content in the introduction has also been modified to clearly express our meaning when we meant to create a combined data set.

Line 138: What is meant by “regional-scale monitors”? Do the authors mean that many samplers operate across the United States?

>> We refer this to regional-scale monitoring stations. This is corrected in the revised version.

Line 139: replace “understanding long-range transport” with “understanding long-term trends”.

>> This is addressed.

Line 139-140: I suggest replacing the Malm 1989 reference with the Malm 1994 reference (Malm, W. C., J. F. Sisler, D. Huffman, R. A. Eldred, and T. A. Cahill (1994), Spatial and seasonal trends in particle concentration and optical extinction in the United States, J. Geophys. Res., 99(D1), 1347-1370)

>> This reference is added.

Line 143-144: The IMPROVE samplers typically sample midnight to midnight, was the sampler at Egbert running on a different schedule?

>> Yes, the IMPROVE samplers at Egbert was run on a different schedule and this has been confirmed by DRI.

Line 148: Are the filters shipped cold?

>> Yes, they are shipped in coolers with ice pack.

Line 155: Spell out CAPMoN.

>> This is addressed.

Line 159: Do the measurements include carbon at all of these sites as well?

>> Historically there have been a number of sites that carry carbon analysis. However, they have been slowly shut down and Egbert is the only site with the longest collection history. This information is included in the revised version.

Line 176: Also see Malm et al. (2001) for a discussion of sampling biases on OC and EC concentrations (Malm, W. C., B. A. Schichtel, and M. L. Pitchford (2011), Uncertainties in PM<sub>2.5</sub> gravimetric and speciation measurements and what we can learn from them, J. Air & Waste Manage. Assoc., 61, 1131-1149, doi:10.1080/10473289.2011.603998.)

>> The reference Malm et al., 2011 is now included.

Line 179: Spell ECCC- Also, please choose notation, either CABM or ECCC. Both are used interchangeably throughout the paper and it is confusing.

>> ECCC is removed from the subtitle.

Line 184: replace "costal" with "coastal"

>> This is addressed.

Line 207: replace "measurements is" with "measurements are"

>> This is addressed.

Line 211: Include "an" between "uses" and "impactor"

>> This is addressed.

Line 212: Replace "Impactor" with "Impactors"

>> This is addressed.

Line 218-222: See the Malm et al., 2011 paper mentioned earlier.

>> The reference Malm et al., 2011 is now included.

Line 228: Can the authors provide some references for the multiple studies?

>> References are now included.

Line 232: Change "introduce" to "introduces"

>> This is addressed.

Line 236: I think you can remove “SRM 8785 & 1649a” from the section header.

>> This is addressed.

Line 246: No correlations are given in Figure 1. Also include figure parts in the text and include OC.

>> Correlations are now included in the figure as well as the text.

Line 248-9: Need figure parts for Figure 2 in the text too (e.g., Figure 2(a)-(d) shows TC, EC, OC, and EC/TC, respectively)

>> This is addressed.

Line 249-250: It is unclear what the authors mean by “Irrespective of data disparity”?

>> This is now removed.

Line 270: I am not sure this is clear: Do the authors just resample the high resolution data for different averaging times? When they say different data sets do they mean the same measurement just with different averaging times? Wouldn't you expect these to compare well? Or do they compare EC to the PSAP measurement? The figures have units of  $Mm^{-1}$ , so it suggests that they either converted EC to absorption coefficient (if so, what absorption efficiency was used?). Please clarify, including figure caption 3 when “comparison of different sets of measurements” from (c) because it is misleading.

>> In this section, we use the 1 min resolution PSAP data (measuring aerosol absorption, assumed dominantly by BC) as a common data set. We then average this data set to the once every third day resolution to simulate IMPROVE and CAPMoN data. We also average the 1 min PSAP data to weekly integrated values to simulate the CABM data. The reason we do not directly compare IMPROVE or CAPMoN data with CABM is because these measurements were not in same sampling frequency and therefore when converting these data to monthly averages, there is no way to know if any difference in monthly means was caused by the natural data variations in the original measurements or it was due to the difference in sampling frequency (it could be caused by both factors). In addition, this analysis was done by Yang et al (2011) and therefore it is not repeated in this manuscript. To ensure no obvious bias caused by the difference in sampling frequency, two different monthly means of PSAP (by every three days vs. by weekly integrated) are directly compared. We have clarify the paragraph in the revised version.

Line 255: Which EC/TC value was further verified? Also, replace “sample” with “samples”

>> In Figure 2d (the original Figure 1d), the three blue bars represent the EC/TC ratio reported by the certificate and also determined from the inter-comparison from the TEA and TOA methods. The green bar represent the EC/TC value calculated from an independent method based on carbon isotope. Here we mean to verify the EC/TC values determined from the TEA/TOA method by carbon isotope method.

Line 283: Over what time period?

>> This is addressed.

Line 288: Can the authors comment on the offset (nonzero intercept) and what it implies in terms of sampling artifacts or biases?

>> A linear regression fit forcing through the origin was applied to Figure 5. The authors believe a fit through zero makes more sense because any non-zero intercept would imply that the

artifact correction obtained from the backup filter was either too much or not enough compared to the actual artifact. The fact that the intercepts were insignificant suggests this is a reasonable assumption and the artifact correction was reasonable.

Line 298: Yes, the POC correction directly influences EC concentrations. Can the authors comment on this vapor adsorption issue with respect to the PSAP weekly comparisons?

>> PSAP in an in-situ instrument that continuously measures the changes in the amount of light transmitted through a quartz filter when particles are deposited onto the filter inside the PSAP. Even though filter media is involved in PSAP measurements, vapor adsorption is not expected to be an issue for PSAP measurements because there is no heating involved, so the adsorbed materials do not char and contribute to absorption.

Line 303: Add a period and start “An optical correction” as a new sentence.

>> This is addressed.

Line 317: Include “monthly mean” before DRI-TOR CAPMon measurements and “comparable to the concentrations derived from the IMPROVE\_A: : :.”

>> This is addressed.

Line 320: What are considered “good correlations”?

>> We consider measurements with correlations above 0.8 to be a good correlation.

Line 351-352: Can the authors describe Figures 7a-c before 7d to keep them in order?

>> This is addressed.

Line 355: At what level of significance?

>> Here the significant correlation is a relative comparison based on the correlation coefficient. We have corrected the wording in the sentence to avoid confusion.

Line 358: I am not convinced the normalized analysis is necessary and adds to the paper. The comparisons between samplers would change depending on what the data are normalized to (choose a different month or an annual mean for example). The comparisons already discussed are more useful because they show the true biases. The diurnal wind cycles on the timelines could be added to the earlier timelines if the authors want to include that analysis.

>> We agree with the referee and we have removed the normalized analysis section and combine some of the information into the section where inter-comparison of the absolute measurements. Because of this, Figure 8 and 9 are now removed from the manuscript.

Line 393: I think elevated carbon concentrations in summer are better shown in Figure 6 given the averaging times.

>> We have modified the sentence to reference this.

Line 413: When are the concentration in the N and NW higher?

>> For OC, elevated concentration could occur during SOA formation when air mass is originated from the N and NW. For EC, elevated concentration could potentially be related to forest fire emissions although more research is needed to verify this.

Line 414: Do the authors mean residential instead of residual?

>> Thank you, and this is addressed.

Line 431: How appropriate is the comparison with ECT9 POC since this is a nonlinear relationship?

>> The authors do not totally understand this comment. However, the corresponding text has been revised to avoid confusion.

Line 447: Also include longer sampling time.

>> This is addressed.

Line 452: What are typical measurement uncertainties? Are these greater?

>> Typical uncertainties could be about 15% for individual OC and EC measurements. The monthly averages should be higher than 20%.

Line 452: Note that others have performed similar comparisons across networks (CSN and IMPROVE) for continental scale integration. Biases for both OC and EC between networks were less than 10% (similar sampling and analytical procedures). Hand, J. L., B. A. Schichtel, M. Pitchford, W. C. Malm, and N. H. Frank (2012a), Seasonal composition of remote and urban fine particulate matter in the United States, J. Geophys. Res., 117, D05209, doi:10.1029/2011JD017122. Hand, J. L., B. A. Schichtel, W. C. Malm, and N. H. Frank (2013), Spatial and temporal trends in PM<sub>2.5</sub> organic and elemental carbon across the United States, Advances in Meteor., 2013, Article ID 367674.

>> The reference has been included accordingly.

Line 504: This link did not work, it needs to be updated:

<http://vista.cira.colostate.edu/Improve/improve-data/>

>> The authors cannot locate the above link. We believe there was a mistake for not copying the proper link. The authors have checked the link

([http://vista.cira.colostate.edu/improve/Data/QA\\_QC/Advisory.htm](http://vista.cira.colostate.edu/improve/Data/QA_QC/Advisory.htm) ) in the acknowledgement section and ensure it is working.

Line 699: Table 1. Can the authors clarify: Is “IMPROVE” under CAPMoN consistent with lines 170-171 that lists IMPROVE-TOT for 2005-2007 and IMPROVE\_TOR protocol? It is challenging to keep these different protocols straight and so careful attention to how they are referred to in the paper and the tables would help.

>> The authors understand the concern from the referee. We have modified the names of the protocols throughout the paper to ensure they are consistent.

Line 702: Table 2: Similar comment, here it is referred to as “Sunset-TOT”. The number of significant digits included in this table seem unnecessary.

>> The protocol name has been verified to be consistent with other parts of the manuscript. We keep the additional significant digits to ensure no round off error when those information will be used by the readers.

Line 710, 713: I don't think these tables are necessary, see earlier comment.

>> This table has now been removed from the main paper and be included in the supplementary information.

Line 718: Figure 1: Again, please be consistent with ECCC and CABM

>> CABM (Canadian Aerosol Basement Measurement) is our network name whereas ECCC (Environment and Climate Change Canada) is our institution name. We believe ECCC is more appropriate in Figure 1 (now become Figure 2) caption.

Line 725: Figure 2, Same comment as previous figure. What is ICP? Please relate x-axis labels to the caption description.

>> This is addressed.

Line 732: Figure 3: See earlier comment- this comparisons is unclear.

>> The corresponding paragraphs have been modified to provide additional information to explain these figures.

Line 740: Figure 4: It would help to see the comparisons in (b) and (c) if the scales were reduced. Again, note the data description in the figures do not match the discussions or tables (e.g., "Sunset-TOT")

>> We have modified the names of the protocols throughout the paper to ensure they are consistent with the description in the Figure caption.

Line 757: Figure 6: Please include location in this figure caption so it is clear that the three different networks are collocated at one site.

>> This is addressed.

Line 766: Figure 7: Which "IMPROVE" are the comparisons made against? Please be clear in the caption to match the axis labels.

>> This is addressed.

Figures 8 and 9: Are unnecessary and do not lend to a better understanding of the comparisons.

>> These figures are now removed.