**Author responses to Referee #3's comments of amt-2019-351 (*Evaluation of equivalent black carbon (EBC) source apportionment using observations from Switzerland between 2008 and 2018*)**

January 29, 2020

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## Response to reviewers

## Referee #3

Grange et al. present an evaluation of source apportionment via the aethalometer model from six Swiss sites. They compare their obtained fossil and wood burning fractions from the model to expected diurnal patterns and ambient temperature. They also compare the aerosol Ångstroem exponents (AAE) they use in the model to the binned AAEs of the full (non-apportioned) time series. Finally, they also show and discuss the trends in fossil/wood burning fractions. This article is informative the figures are well designed and illustrative. However, there are some important issues regarding the data processing, at least as it is outlined in the text. I also note that the technical aspects of the paper are not as in depth as work by e.g. (Fuller et al., 2014) or (Zotter et al., 2017) who compare their data to external tracers, such that an experienced user of aethalometer data might learn more from the reported trends and source apportionment than the technical aspects presented. Nevertheless I recommend publication after the following issues are addressed.

Thank you. We have addressed all of the referee's comments below.

### Major issues

- Most data in the paper are from the Swiss National Air Pollution Monitoring Network (NABEL). I tried to access this site using the link in the references, but found that the link was no longer active, whereas the authors state that data are available as described in the text. Furthermore there is no description of data quality control/assurance. Are all data aggregated to hourly resolution or are some data points removed due to instrumental issues etc.? If points are removed, what is the threshold at which an hourly data point is still reported (i.e. what number of missing points are allowed?). The authors should update the information they provide on data availability and describe the quality control measures. Payerne and Rigi are EMEP sites and aethalometer data are publicly available at ebas.nilu.no.

We thank the referee for reporting these data accessibility issues. In response to this, we have deposited the absorption observations, site metadata, and instrument locations data used in the data analysis in a persistent and publicly accessible data repository (`https://doi.org/10.5281/zenodo.3626658`). The data section now contains more information on data quality control and assurance procedures with a new citation which outlines these operations in detail.

- The authors do not seem to have access to time series at the original time resolution of the instruments since they write 'Generally, the observations were stored as hourly means, but for the data which was at higher resolution (10 and 30 minute means)', implying that raw instrument output was not available? Checking these time series is important for several reasons, e.g. to ensure correct application of the automatic loading compensation in the AE33 instruments (i.e. there should be no structure in the absorption or EBC time series related to tape advances), or that no short term spikes are present in the data which could bias the averaging. Plotting the AAE over time is also useful. This lack of information weakens the arguments the authors make regarding the failure of the aethalometer model apparent at the San Vittore site. In the text they assert that this failure is due to the presence of a third source (fresh wood smoke). Here it is important to rule out other causes of failure, which might arise due to errors in the data processing (e.g. incorrect loading compensation). If possible, time series should be shown in the annex. If not, this weakness should be mentioned in the text.

  We have conducted our data analysis on observations which have been subjected to quality control and assurance procedures (see the response above) and therefore these data are not "raw" data off the instruments. We would argue that the use of such data is more relevant to the research community because such data are generally what is accessible to the public. We do however accept that using the raw instrument outputs may offer some additional value for testing and checking, it does not weaken our conclusions or results.
  We have added a paragraph further detailing the data processing and quality control and assurance procedures. The paragraph reads: "The standard procedure for absorption monitoring data was for 10 and 30 minute means to be calculated from one minute observations

which were logged on-site directly from the instrument. All aggregations required a data capture threshold of 60 % for a valid summary to be produced. Additional quality control and assurance procedures were undertaken quarterly including: cleaning of the inlet, leak testing, and cleaning of the analytical zone of the instruments. The responses of the AE33 instruments were also checked with optical reference filters regularly. The data were ratified on a monthly basis and compared across the different measurement sites along with other air pollutants and suspicious measurements were invalidated. The raw instrument outputs are archived, but are not routinely used in the data processing within the NABEL monitoring network. However, the raw data can be consulted in case of questions concerning data quality."

A time series of $\alpha$ has also been included (Fig. A4 and below) to address this, and other comments.

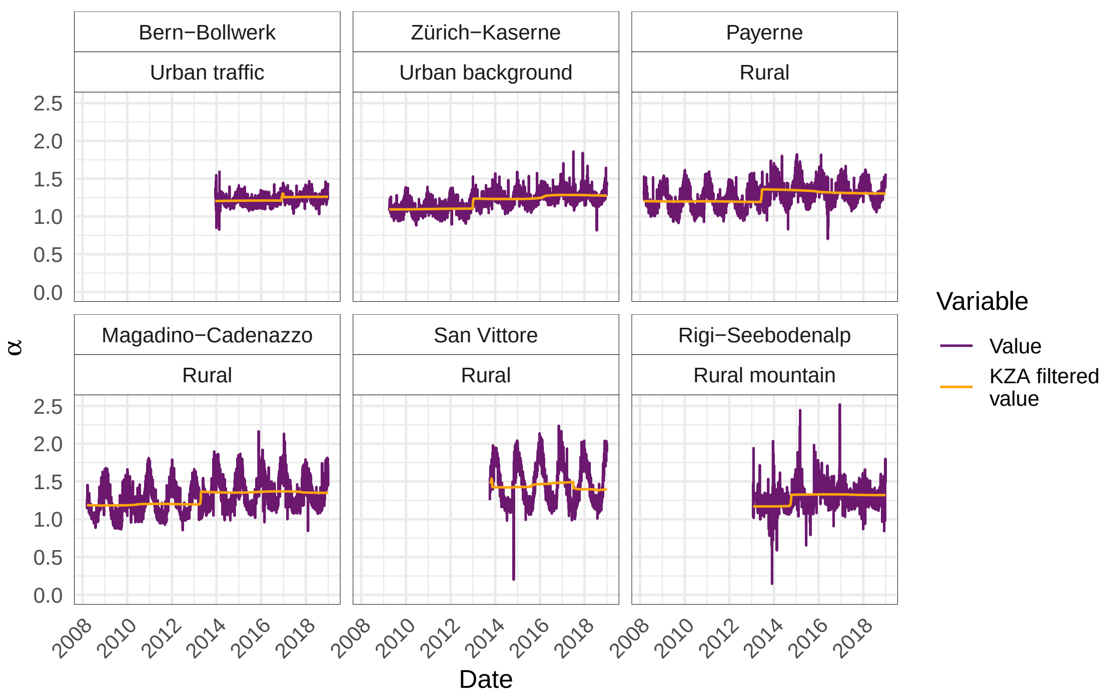


Figure 1: Time series of daily and adaptive Kolmogorov-Zurbenko filtered (KZA) Ångström exponents ($\alpha$) for six equivalent black carbon (EBC) monitoring sites' absorption observations in Switzerland between 2008 and 2018. $\alpha$ has been calculated by wavelengths between 370 and 950 nm.

### Minor comments

- Change 'appointment' in the title to 'apportionment'.

  Done.

- Pg 1, ln 16: change 'has be' to 'has been'

  Done.

- Pg 1, ln 17: PM2.5 is undefined

  $PM_{2.5}$ is now defined in the abstract and the first occurrence in text.

- Pg 2, ln 19: Suggest using 'direct legal limits' since in the unlikely event that BC is itself above the limit values for PM2.5 then BC would be above the legal limit. Non-binding provisions for BC limits are also included in the Gothenburg protocol, see also (Shapovalova, 2016).

  The text now reads "...direct legal limits...".

- Pg 5, ln 12: Data from AE31 instruments are included and the authors write 'All absorption observations had been compensated for the filter loading and shadowing effects with the instrument model's respective algorithms before this analysis was undertaken'. However, the output from the AE31 is slightly different to that of the AE33 in that a correction factor for loading is not automatically applied by the instrument and must be done in post processing. Please clarify.

  This text has been altered to clarify the differences between the two instrument models: "The differences between the AE31 and AE33 technologies mean that the algorithms which compensate or correct for filter shadowing effects and filter loading effects are different. Notably, data from AE31 must be compensated for such effects with a post-processing procedure while the AE33's algorithms are conducted on-board as part of the measurement cycle. For the full description of the compensation procedures, see...".

- Pg 7, ln 9: The authors write 'When using aethalometer and EC data together, the aethalometer observations were aggregated (as arithmetic means) to daily 10 resolution'. I assume the data were averaged to the start and end times of the filters? In which case it would be better to write something like 'data were window averaged to the start and end times of the filter sampling'.

This point was also brought up by Referee #2 and has been addressed. The sentence is now much clearer and describes the data handling step in detail: "When using aethalometer and EC data together, the aethalometer observations were aggregated (as arithmetic means) to daily resolution (midnight to midnight) to ensure the observations spanned the same time period and duration. Only days with both EC and absorption observations were used for these comparisons because interpolation of the less frequent EC data was not attempted.".

- Pg 7, ln 23: Use 'recommended by' instead of 'reported by'.

  Done.

- Pg 8, ln 12: What was the least squares algorithm used? Given that there is uncertainty in both x and y, orthogonal distance regression should be used.

  We used simple least squares regression models. Although we accept that uncertainty in the EC observations ($x$) could be argued, the EN16909 thermal method used to determine EC is a reference method and can also be considered "truth". The very high degree of correlation between EBC and absorption would also result in the different estimators producing very similar results. We believe the use of total least squares/orthogonal distance regression is a preference and consider our approach valid for our analysis.

- Pg 8, ln 14: Here it would be better to use 'an average of', rather than a 'range of' since the range is later given as 3 m2/g. i.e. write an average of 10 +- 3 m2/g

  The sentence has been altered and now reads: "The sites' MAC values at 950 nm had an average of $11.3 \pm 2.9\,\mathrm{m^2\,g^{-1}}$ during the analysis period..." Please note that the discussion on the MAC values has been expanded due to the comments of Referee #2, point 13.

- Pg 8, ln 18. The report by Anonymous Referee #2 discusses the issue of the large variability in MAC. How much of the variation in MAC is actually due to instrument differences? It is natural to expect some variability due to this. The effect of this will have been somewhat obscured by using the rolling average presented in the paper, so that Fig. A1 likely does not show the full magnitude of the effect. It would be informative to include e.g. seasonal MAC values on an instrument by instrument basis e.g. for each line of Table 2. A version of Fig. 3 highlighting instrument shifts would also be informative.

  A table containing mean MAC values by site, season, and instrument was produced but it is not practical to include in the manuscript due to the length of the table (154 rows).

Therefore, a supplementary figure (Fig. A2 and below) has been added showing these data. Fig. 3 (also below) has also been enhanced to show the times when instruments were changed as requested. Please note that the discussion on the MAC values has been expanded due to the comments of Referee #2, point 13.

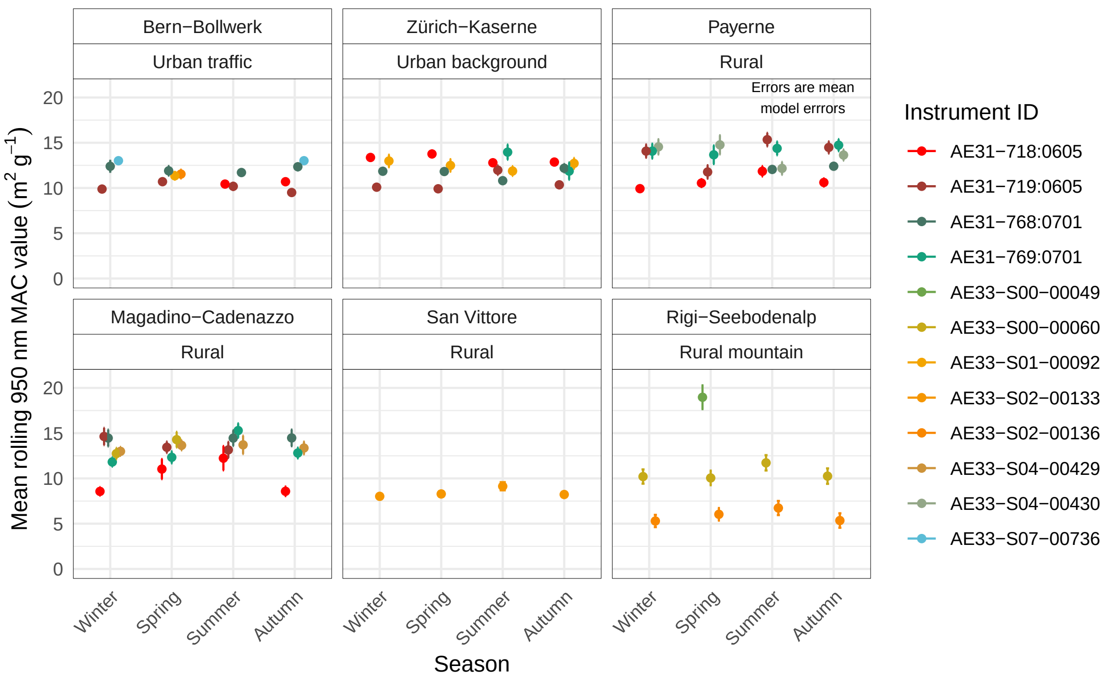


Figure 2: Mean seasonal and instrumental mass absorption cross section/mass absorption coefficients (MAC) for 950 nm for six equivalent black carbon (EBC) monitoring sites in Switzerland between 2008 and 2018.

- Pg 8, ln 27: change 'is' to 'are'.

  Done.

- Pg 14, ln 12: change 'negative' to 'negatively'

  Done.

- Pg 22, Figure A2 caption: Please change the caption to reflect that these are the steps used in the paper and not a general requirement, e.g. to 'A flow chart of the data processing steps used to apply the aethalometer model'. Some of the steps could have been done in a different order, and apportionment of absorption coefficients is also possible without conversion to EBC.

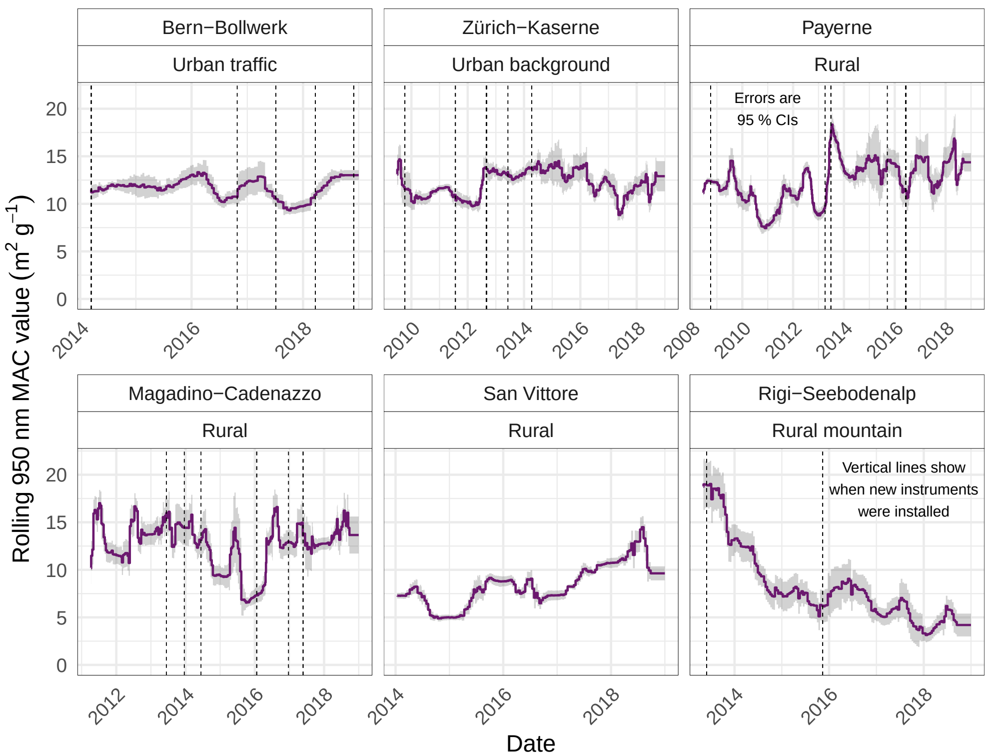


Figure 3: Mass absorption cross section (MAC) coefficients for 950 nm for different monitoring sites as calculated by rolling simple linear regression models with windows of 180 days (alignment of the window was the centre of the period).

Done.

## References

Fuller, G. W., Tremper, A. H., Baker, T. D., Yttri, K. E., and Butterfield, D. J. A.: Contribution of wood burning to PM10 in London, 87, 87-94, 2014.

Shapovalova, D.: The Effectiveness of Current Regulatory Models of Gas Flaring in Light of Black Carbon Emissions Reduction in the Arctic, in: Global Challenges in the Arctic Region: Sovereignty, Environment, and Geopolitical Balance, Routledge, 325-344, 2016.

Zotter, P., Herich, H., Gysel, M., El-Haddad, I., Zhang, Y., Močnik, G., Hüglin, C., Baltensperger, U., Szidat, S., Prévôt, A. S. J. A. Evaluation of the absorption Ångström exponents for traffic and wood burning in the Aethalometer-based source apportionment using radiocarbon measurements of ambient aerosol, 17, 4229-4249, 2017