Author responses to Referee #2's comments of amt-2019-351 (*Evaluation of equivalent black carbon (EBC) source apportionment using observations from Switzerland between 2008 and 2018*)

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Response to reviewers

Referee #2

This manuscript reports trends in equivalent black carbon (EBC) at several sites in Switzerland. Using aethalometer data, the authors apply a model to apportion EBC to traffic and wood burning sources that requires using Angstrom exponents and the assumed spectral dependence of wood smoke and traffic emissions. The data were interpreted in the context of these sources, including seasonality and diurnal patterns. The authors also report analysis of long-term trends in both the traffic and wood burning fractions. Results corresponding to periods when the aethalometer model failed provided additional feedback for when it is unable to successfully apportion sources, in this case in situations with freshly emitted wood smoke. The paper is clearly written and organized and the methods are well described and sound. The results are helpful in understanding the role of emission mitigation strategies for traffic and biomass burning sources. I recommend publication after addressing comments below.

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

1. P1, Line 7: Given the issues the model had with what appeared to be fresh smoke, would it be fair to refer to this fraction as aged?

   This point is addressed below in point 2.

2. P1, Line 12: This goes back to the previous comment. To clarify, calling the smoke fraction "aged woodburning" would help distinguish these sources.

   These two above points can be addressed together. The woodburning fraction which the aethalometer model apportions is considered a mixture of fresh and somewhat aged woodburning sourced BC. In a typical urban area, such as Zürich, the woodburning sourced BC

encountered will be a combination of aged particles as well as freshly emitted emissions. We believe the model failure shown in San Vittore was caused by a much greater proportion of fresh woodburing emissions which has different spectral characteristics than the more commonly encountered mixed woodburning BC. Therefore, calling the woodburning fraction 'aged' does not seem to be truly correct here.

3. P1, Line 15: Change "deceases" to "decreases"

   Done.

4. P1, Line 16: What does "This" refer to at the beginning of the sentence?

   The sentence now begins with "This lack of reduction in EBC_{WB}..."

5. P1, Line 17: If the site is a likely representative location and EBCWB has not decreased, but the other sites have, how is it representative of ineffective controls on wood burning? Are the same management controls applied everywhere?

   This site (Magadino-Cadenazzo) represents the environment south of the Alps and is distinct when compared to the monitoring sites on the Swiss plateau. We believe Magadino-Cadenazzo is a very good representative site for such environments.

6. P2, Line 6: Change "fuelled" to "fueled"

   Fuelled is the preferred British English spelling which this manuscript has been prepared in so this has not been changed.

7. P7, Line 2: Change "consistently" to "consistent"

   Done.

8. P7, Line 8: What period was considered a daily sample? Midnight-midnight?

   The daily averages were indeed midnight to midnight. The text now reads: '...to daily resolution (midnight to midnight)...'

9. P7, Line 10: How were EC data applied with this frequency since BC and EC would overlap only on certain days? Was the EC sampling schedule the same at all sites?

   Extra text has been added to clarify this point: "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."

The EC sampling was very similar among the sites with minor variations to sampling frequencies over the analysis period.

10. P7, Line 11: How was PM2.5 measured at all of the sites? What sampling frequency?

    This sentence has been changed to make the type of $PM_{2.5}$ observations clear: "Only daily validated data were kept for analysis with most of the observations being sourced from high volume samplers."

11. Page 7, Line 16: Change "measures" to "data"

    Done.

12. Page 8, Line 4: Were any trend analyses performed on the EC data to test whether EC trends generally followed the overall BC trends?

    Trend analysis of EC has not been conducted as part of this work. However, this is a worthy idea for future work but may be challenging due to less frequent observations before about 2010.

13. Page 8, Line 16: Where was this seasonal pattern observed? At all sites? A range of 3 m2/g is very large, can the authors comment on the physical reasons why the MAC would vary this much on a seasonal basis? They also vary considerably from site to site. Were EC data examined on a site by site and seasonal basis? Do they show this much variability?

    The MAC value discussion has been expanded and now includes a supporting figure (Fig. A2 and below) which shows the seasonal MAC values by site and instrument. The text now discusses that the seasonal cycle was observed in all locations: "The range of the seasonality was as low as $0.4\,m^2\,g^{-1}$ at Zürich-Kaserne, but as high as $2.3\,m^2\,g^{-1}$ at Magadino-Cadenazzo. The seasonal variation was also accompanied by intra-instrumental variation."

    The MAC value variation may be due to the woodburning emission source being 'switched on and off' during the year which would substantially alter the seasonal aerosol composition. However, this is not explicitly investigated here and do not wish to speculate on the causes. We point to Zotter et al. 2007 (https://www.atmos-chem-phys.net/17/4229/2017/) where changes in MAC values are discussed.

    The EC observations were checked for seasonality and their range was not as great as 3

units, but showed substantial variation with winter being the most polluted season due to a combination of emission strength and meteorological controls.

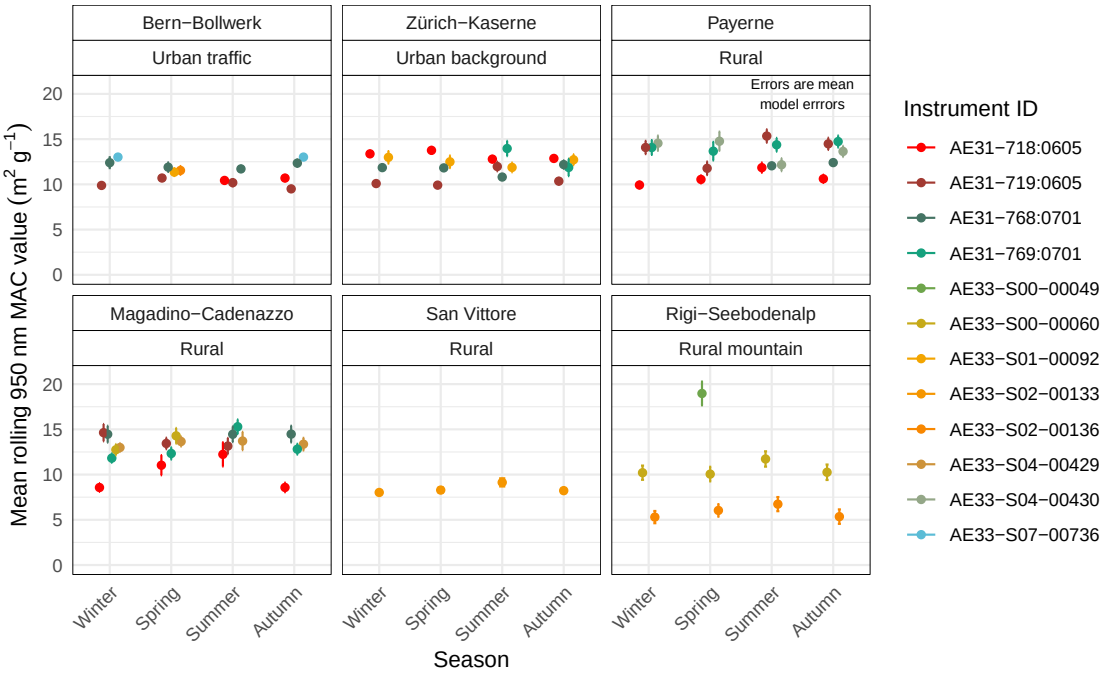

Figure 1: Mean seasonal and instrumental mass absorption cross section/mass absorption coefficients (MAC) for 950 nm for six equivalent black carbon (EBC) monitoring sites in Switzerland between 2008 and 2018.

14. Page 9, Line 6: Can the authors provide a brief explanation on how Angstrom exponents were calculated? Are they ratio of two wavelengths or a fit of all wavelengths?

    A sentence has been added explaining the calculation process: "Here, α was calculated by curve fitting all absorption wavelengths (370, 470, 520, 590, 660, 880, and 950 nm) with exponential regression models."

15. Page 10, Line 2, Figure 4 caption. Can the authors add the wavelength range for the Angstrom exponents?

    The caption of the figure now reads: "Counts of binned Ångström exponents (α) for three equivalent black carbon (EBC) monitoring sites' hourly absorption observations in Switzerland between 2014 and 2018. α has been calculated by seven wavelengths between 370 and 950 nm."

16. Page 12, Line 4: What about wood burning as local emissions?

This monitoring site would be influenced by local woodburning and other emissions, but based on the data, this influence is very small. The sentence states this clearly: "This monitoring site is located at an elevation of 1031 metres, is isolated from significant local emissions, and is intermittently in the boundary layer and therefore, at times is not influenced by surface source activities."

17. Page 12, Line 9: Why would this feature be stronger for traffic than wood burning sources?

    Although this feature is discussed later in the manuscript, this sentence has been expanded: " This feature was clearer for EBC_{TR} than EBC_{WB} suggesting traffic, rather than woodburning emissions are more influential at Rigi-Seebodenalp."

18. Page 14: Line 23: It appears from the figures that a couple of sites do not have data before 2013. Trend analyses performed over this short of period can be misleading when compared to sites with longer periods.

    The EBC monitoring sites do have different start dates which creates a situation where the trend analysis is potentially weakened, these are the data which exist and little can be done to address this.

19. Page 14: Line 26: The Payerne site appears to have a break around 2014, after which the data are relatively flat. A similar pattern may occur at Magadin-Cadenazzo and Zurich sites. Is this an instrument artefact?

    We have investigated these potential breakpoints thoroughly. We have added a supplementary figure (Fig. A4 and below) and a paragraph in the trends section explaining these instrument artefacts. The paragraph reads:
    "EBC_{TR} trends at Payerne, Magadino-Cadenazzo, and potentially Zürich-Kaserne suggest that breakpoints in the observations are present at the middle of 2013 (Fig. 9). Curiously, these changes were not observable in the absorption observations themselves, but could be detected in the calculated α with adaptive Kolmogorov-Zurbenko filters (KZA filters) (Fig.A4). The times of these small breakpoints could not be robustly traced to operational activities, but they might have been caused by different batches of aethalometer filter-tape which were continuously introduced across the monitoring network at this time. Therefore, these data suggest that these breakpoints are instrument artefacts. The identification of these features reinforces that the use of the aethalometer model is very useful, it is a pragmatic

technique which requires careful evaluation."

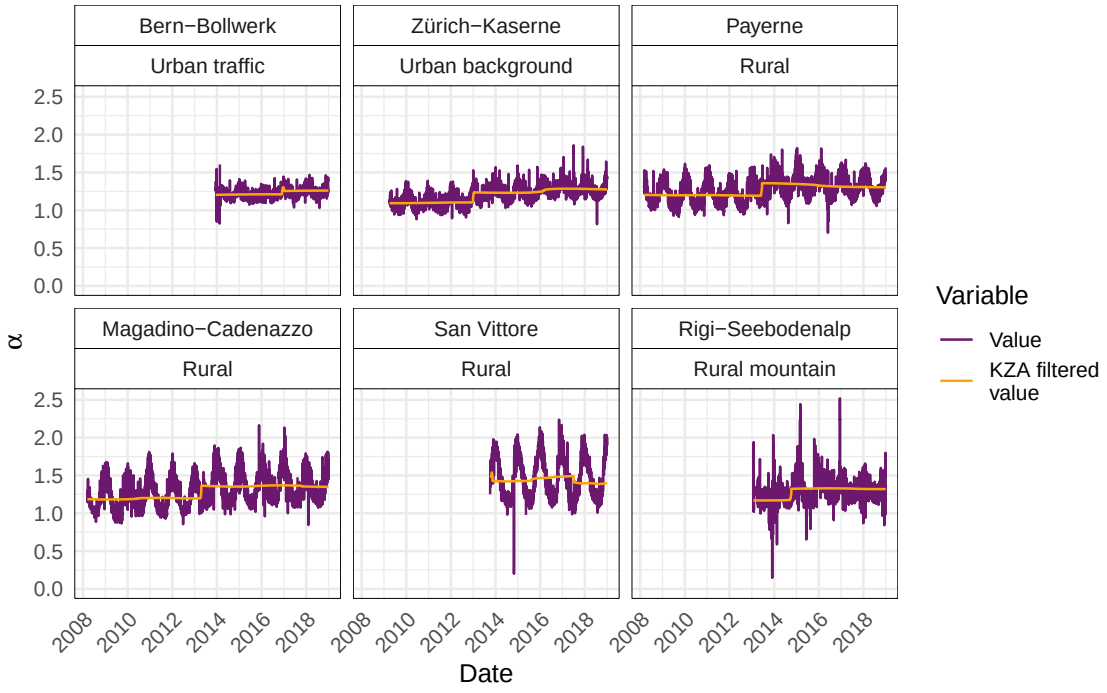

Figure 2: Time series of daily and adaptive Kolmogorov-Zurbenko filtered (KZA) Ångström exponents (α) for six equivalent black carbon (EBC) monitoring sites' absorption observations in Switzerland between 2008 and 2018. α has been calculated by wavelengths between 370 and 950 nm.

20. Page 15: Line 12-13: But the time period is much shorter and during this period, other sites had flat trends (Payerme and Magadino), so it may not be fully reflective of what is happening over the longer time period.

    We have conducted to the trend analysis on the observations which are available. We accept that the non-uniform data spans is a limitation of the study, but this cannot be changed due to the variations of the observational record and believe the trends are representative of the site types discussed.

21. Page 16, Line 5: Over the same years?

    The EBC/$PM_{2.5}$ ratio analysis is conducted across the same period. The sentence has been modified to make this clear: "The seasonal EBC/$PM_{2.5}$ ratios for the five Swiss EBC monitoring sites between 2014 and 2018 are shown..."