

Dear editor,

Please find our responses to the referees on ACPD discussion section. I also attached a revised version, where the revised texts are in red font.

Thanks

Dantong Liu

We thank both referees' comments and suggestions, here are our responses. The referees' comments are in *Italic*, our responses are in normal font.

R. M. Healy's comments:

This article should be suitable for publication subject to some minor revisions as outlined below.

General comments:

Title: I would suggest removing the letter 's' after apportionment and aerosol. Also, were the measurements not performed in both summer and winter?

The title will be revised. Although the experiments were performed in both winter and summer, the winter results are the main focus of this study and the summer is only used to aid the analysis of winter data.

Was an SP-AMS available during the Clearflo campaign? It seems like a comparison between SP2 apportionment and SP-AMS full mass spectra or PMF factors would be relevant and useful here.

While a comparison with the SP-AMS would be relevant, the instrument was operated in the non-standard 'vaporiser in' mode of operation, which introduces complications in the data analysis and detract from the main focus of this work. This will be discussed in a future publication.

It is confusing at times whether the summer data are analysed here or not. For example in Fig. 6 they are included. Why refer to these data as a separate study throughout?

As the response above, the summer data is not the focus of this study, such as in Fig.6 the BC size distribution data in summer is only used to support a conclusion for winter data that in winter the BC are in a state of multiple sources.

Figure 3: middle and bottom panels appear to have different air mass origin assignments at times.

This will be corrected in the revised version.

Section 3.3: last paragraph needs to be rewritten as it is confusing in its current form. There is nothing wrong with the approach but the impact is reduced.

This will be rewritten in the revised version.

Page 16318, line 20-25: I don't think the relative importance of local sources and processing can be assessed here.

This part will be rewritten in the revised version.

Conclusions: Rewrite last paragraph to improve grammar and impact.

It will be rewritten in the revised version.

Specific comments:

Page 16293: line 22, replace 'or' with 'and' before easterly

Page 16294: line 1-2, replace hyphens with words to separate the air masses and value ranges, it is quite confusing in the current style.

16294, line 25: should be 'targeted'

16296, line 6: fix multi wavelength

16299, line 10: What is NK?

16299 and References: Best not to give a publication year for articles not reviewed or published yet. Simply 'in preparation' is fine

16302, line 3, should be 'beyond the scope'

16303, line 17, add 'countries' after Benelux

16305, line 1-5 is stated as a fact but should be rephrased as a hypothesis

16306, line 18: should be 'given that'

16307, line 24: replace sector with factor

16312, lines 1-3. Removing this line would be better as whether or not this compensation is happening cannot be determined

16312, line 4, mean mass median?

16312, line 6: Is the standard deviation really this low (around 1%)?

The deviation here is reported as geometric standard deviation rather than the normal standard deviation given the former value is more widely used by the modellers when constraining the size distribution.

16312, line 23: receptor site?

16313, line 9-14: this sentence is difficult to follow

16314, line 4: Hyphenation makes this confusing

16319, line 5: Traffic-related?

Figure 6, the error bars are +_, no negative are shown. And the geometric mean is a factor of 10 higher than the median? Or maybe this is the standard deviation of the mass value at this diameter?

This will be clarified in the revised version.

All of the above specific comments will be addressed in the revised version.

D. Baumgardner's comments:

There are several places in the manuscript where I felt some clarity was needed and my comments may be found in the annotated file that I have uploaded with this review.

All of the comments in the attached file will be addressed in the revised version.

There is one suggestion that I will make that the authors are free to ignore or test at some other point. An effective way to extract even more information from complementary measures of size distribution and angstrom exponent is to calculate absorption coefficients from the SP2 size distributions of rBC and a subsequent angstrom exponent. It may be that the size range of the SP2 is too limited to extract an AE that can be meaningfully compared to that from the Aethalometer. On the other hand, closure of this type helps to better understand the sources and might also shed light on the

source of the background that at times is quite large.

As I said, the paper is certainly complete as it is. The closure study would just be an extra nugget of information that I don't believe anyone else has attempted yet.

We thank referee's very inspiring comments and we are preparing a manuscript specifically focusing on the BC optical properties, which will fully include AAE calculation as referee suggests here.