Atmos. Chem. Phys. Discuss., 9, C771–C779, 2009
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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Particle number size distributions in urban air before and after volatilisation" *by* W. Birmili et al.

W. Birmili et al.

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It somewhat disturbs me in this text that formulas and equations are clearly avoided even though they would make the presentation clearer and unambiguous. You could easily add a few formulas there.

Reply: Thank you for this comment. We don't generally avoid equations, but just thought to increase the readibility of the article. In fact, we are happy to include some formulae on the autocorrelation analysis, and the MAAP/aethalometer comparison (see both below).

The MAAP and the aethalometer comparison is reported inadequately by only giving the offset, correlation coefficient and averages. Rather give the linear fit as BC(aeth) =

k × *BC(MAAP) + BC0,* R^2 *= xx.xx*

The exact relations are:

$$\mathrm{BC}_{aeth} = 0.86 \cdot \mathrm{BC}_{MAAP} + 0.24; R^2 = 0.96 \quad (1)$$

and

$$\mathrm{BC}_{MAAP} = 1.11 \cdot \mathrm{BC}_{aeth} - 0.18; R^2 = 0.96. \quad (2)$$

The full cloud of data points is, for your information, plotted in the three Figures attached to this document. It is worth mentioning that for low concentrations ($< 2\,\mu$g per m^3) a squared relationship would represent a better fit. This behaviour is illustrated in Figure 3. We are not aware of the exact reasons for this behaviour.

The MAAP is [referenced as] Sheridan et al., 2005. The proper reference to MAAP is not Sheridan et al. but Petzold & Schoenlinner, J. Aer. Sci 2004 - look for the full ref.

Reply: Thank you for this hint. We will certainly add the original Petzold & Schoenlinner reference. Our intention to include the Sheridan reference was the mention of the Reno workshops, where the MAAP was shown to be equivalent to a selection of other reference methods for aerosol absorption. The Sheridan article reads, however, unfortunate in that the MAAP was described there sometimes under its old name “Carusso’. Meanwhile we have found another reference Petzold et al. (2005), Evaluation of multiangle absorption photometry for measuring aerosol light absorption, Aerosol Science and Technology, 39:40–51, 2005, which we plan to use as a replacement for Sheridan et al.

2.4 “This method was originally developed for the analysis of the hygroscopic growth of particle size distributions (Birmili et al., 2009) but has since been applied to quantify the loss in particle diameter after passing through a TD (Engler et al., 2007)” There is some logical error in this.

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Reply: We developed the “diameter shrinking factor” method in ca. 2004, but the detailed publication of the method has been pending until 2009. Meanwhile, Engler et al. (2007) used that method but gave only a very brief account on the numerical details. As a solution we suggest to reformulate the sentence as “The method was developed to quantify size-segregated diameter changes in particle size distributions that are brought about, for instance, by thermodesorption (Engler et al., 2007), or humidification (Birmili et al., 2009). A detailed account on the numerical procedure is given in the latter reference.”

3.1.4 Auto-correlation analysis The whole procedure is kind of intuitively clear to me but still not exactly. Especially when it goes to the next phase, the factors Fa – Fd and the integrals of these. This all should be explained more in detail, including a couple of formulas. Actually this seems to be an interesting and useful way of handling data — has it been used by others? If yes, give references. If not, the more there is reason to really give the formulas starting really from the definition of ACF.

Reply: The auto-correlation is the cross-correlation of a signal with itself. It is a numerical tool to find repeating (i.e. periodical) patterns in a signal (here: time series of number concentration) that may be obscured by other confounding noise variables. Auto-correlation analysis is widely used in electronic signal processing and has, to our observation, also been used in a few textbooks of environmental statistics. It has, however, found surprisingly few applications in atmospheric applications. Examples are Wigley et al. (1998), Anthropogenic influence on the autocorrelation structure of hemispheric-mean temperatures, Science, 282: 1676-1679 for climatological time series of ambient temperatures, or Wehner and Wiedensohler (2003) Long term measurements of submicrometer urban aerosols: Statistical analysis for correlations with meteorological conditions and trace gases, Atmos. Chem. Phys., 3, 867–879, 2003 and Heintzenberg et al. (2004) Structure, variability and persistence of the submicrometre marine aerosol. Tellus 56B: 357–367 for urban and marine aerosols, respectively. A more recent use of the ACF can be found in Sunder Raman et al. (2008)

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Carbonaceous aerosol at two rural locations in New York State: Characterization and behavior, J. Geophys. Res., 113, D12202, doi:10.1029/2007JD009281.

In conclusion we will add the defining formula for the ACF:

$$r_{xx}(\tau) = \frac{1}{n} \sum_{t=0}^{n} x(t) \cdot x(t+\tau), \tag{3}$$

With $x(t)$ being the standardised time series (zero mean, unit standard deviation) of number concentration here.

We will also and some of the above references plus additional textbook references.

While the ACF yields somewhat "intuitive" results, we are aware of the current constraints of the interpretations of F_a–F_d, especially if they are to be quantitative, for instance, when comparing weekly (7-day) and diurnal (1-day) periodicities. This has also been mentioned by Referee No. 1. For this reason we plan to carry out a brief sensitivity study that is based on synthetic time series, which we will hopefully include in the final version of the manuscript.

"Another explanation, which is hard to verify, could be the presence of absorbing aerosol other than soot during the summer period" The latter is plausible. The aethalometer inlet cutoff was 2.5 μm and your annual cycle showed a peak of 2 μm particles in summer. Soil dust particles may easily contain some light absorbing material that the aethalometer reports as BC.

Reply: Thank you. We will happily strengthen this conclusion in the revised text version.

Figure 11. Bad selection of colors: at least my eyes can hardly distinguish spring and winter dots. Actually I suggest you would make a figure 11B where the same data would be presented as with a average squares and some ranges, for instance 99th percentiles in both x and y directions for all 4 seasons.

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Reply: Thank you. We will pick up your suggestion.

Fig 12. Messy, too many diameters in one rose. Why don't you select only the diameters that you used earlier for instance in fig 5?

Reply: We agree, and will concentrate on the selection of particle diameters displayed in Fig. 5.

Figure 14: Missing the subfigure letters in the text.

Reply: Thank you — this will be corrected.

All the figures associated with the trajectory cluster analysis are messy, there are not clear differences between the various descriptive parameters, for instance size distributions associated with many of the clusters. The main reason seems to be that there are so many trajectory clusters and some of them are actually not very different from each other. I would suggest reducing the number of clusters and redoing the clustering – or if this is too big a work at least reduce the number of size distributions etc. presented in the figures and explain in the text that the size distributions associated with the clusters nn-nn were statistically not different. Just do something to make the figs 14a-c clearer.

Reply: Instead of using 13 clusters, we had previously conducted an analysis using only 7 meteorological clusters. (These two results were, in turn, the most self-contained and easily interpretable solutions of many more (ca. 500) mathematical solutions that were obtained for the cluster problem.) The solution for $n = 7$ has the disadvantage that the main effect leading to high concentration in Augsburg (vertical stratification) appeared only in obliterated shape. The solution using 13 clusters, however, makes this effect transparent. 13 clusters were therefore considered necessary to resolve the many occurring weather situations in terms of the variability of 3D-trajectories and vertical temperature profiles.

As a solution we are happy to pick up your suggestion to simplify educe the number of

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size distributions presented in the Figures. Instead of all single lines we think of plotting the essential outstanding clusters 4, 8, and 12, and for all others only the range in which the corresponding distributions reside.

Wolfram Birmili, on behalf of all co-authors.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 9171, 2009.

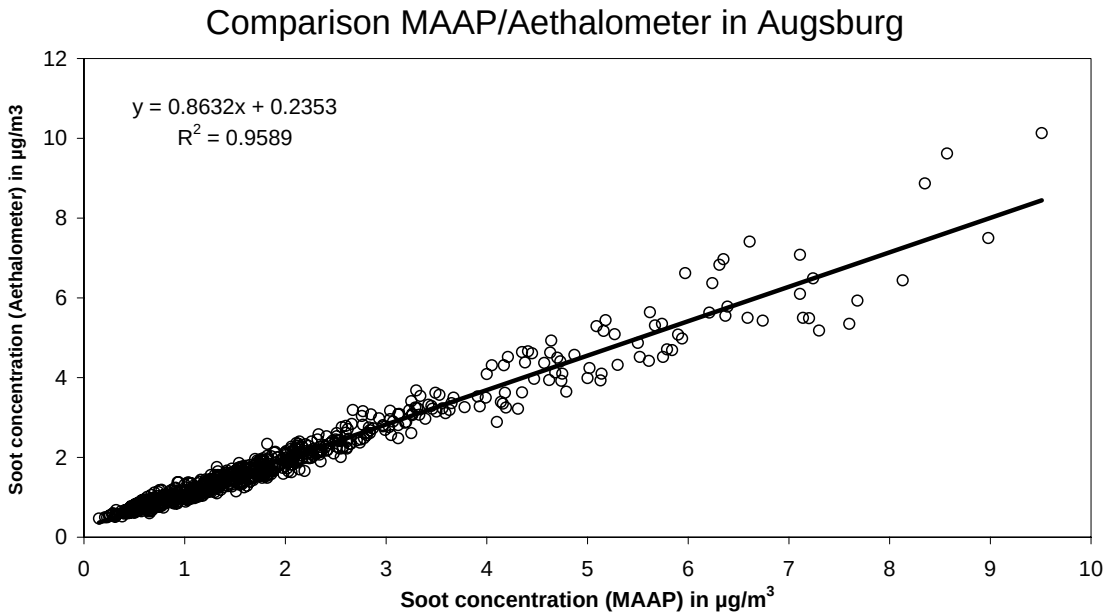

Fig. 1.

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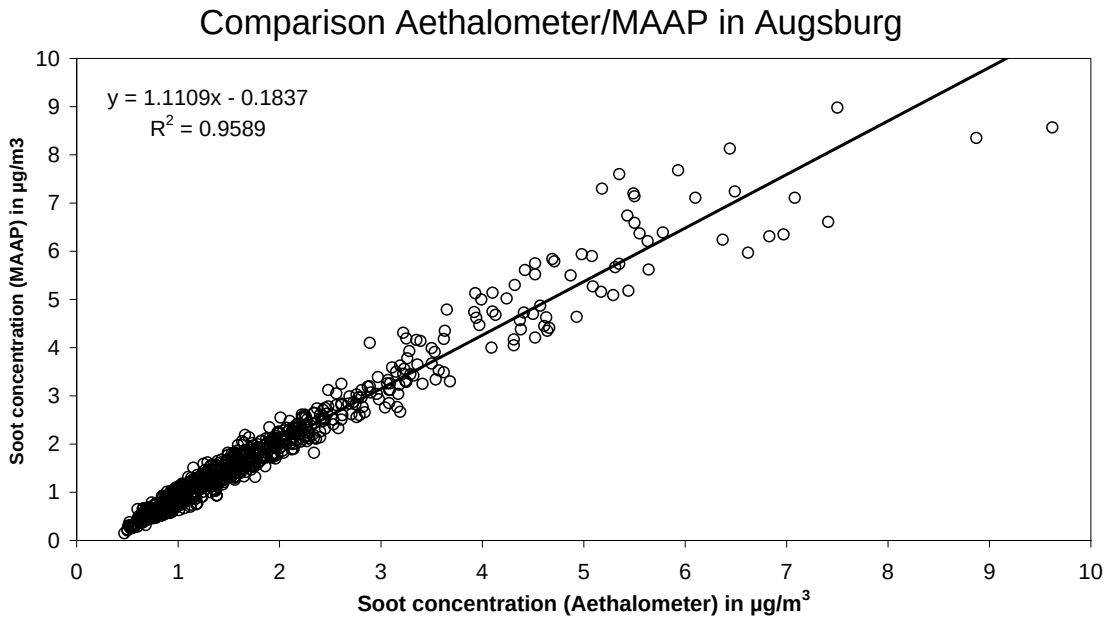

Fig. 2.

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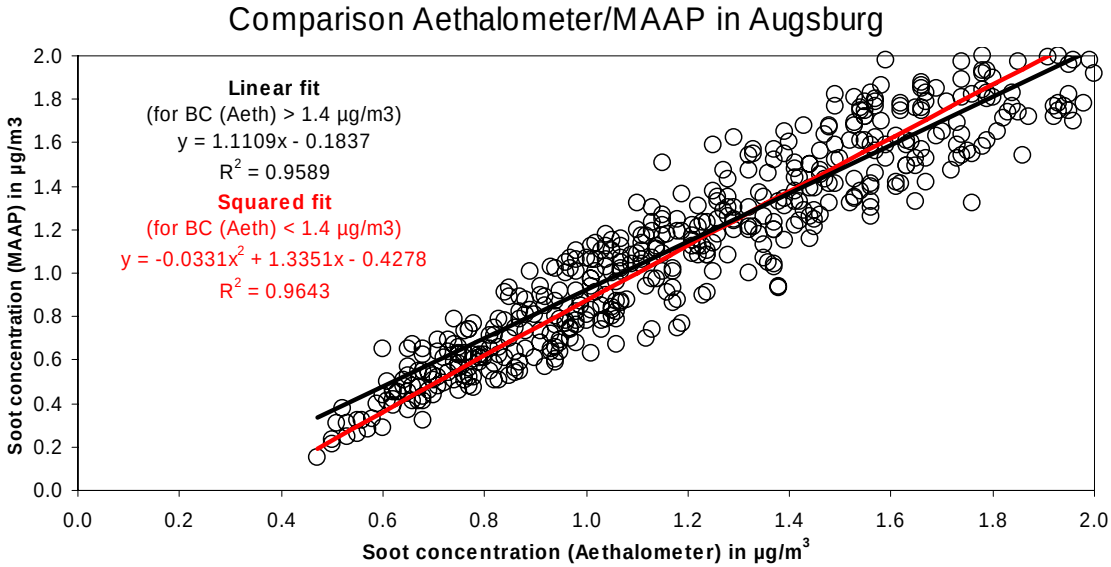

Fig. 3.

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