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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Size distributions of non-volatile particle residuals (D_p<800 nm) at a rural site in Germany and relation to airmass origin" *by* C. Engler et al.

C. Engler et al.

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1 General comments

1. "Section headings..."
   Results presented in section "Methodology" do not belong to the results of this study, which is the reason, why they are not presented in the "Results" section, which is named "Effects of air mass history" in this work. They are only shown to demonstrate the used method in greater detail.

2. "Please add quantitatively..."
   As recommended by the referee, a selection of quantitative multimodal lognormal

size distribution parameters was added for all ambient and non-volatile size distributions. The parameters included the number concentration, geometric standard deviation and mean geometric diameter for each of the three lognormal modes accumulation, Aitken, and nucleation mode. The lognormal modes were fitted using a Levenberg Marquardt least squares fitting routine, which was also used to fit size distributions in an earlier paper (Birmili et al., 2001).

3. "The justification of the applied data analysis scheme..."
   The question after the maximum number concentration is not mandatory, since the peak-height is not equal to the (integrated) number concentration in a certain size range. In a certain size range there could be a large number of non-volatile particle residuals present, but theses residuals could partly grow under ambient conditions. This would result in a broader distribution with lower maximum number concentration in the non-conditioned state. But after the thermal conditioning, the number size distribution would be narrow again with a higher maximum number concentration. However, in this case, there is no new particle formation in the thermodenuder!

4. "In Figures 8 and 9..."
   See point 3. It is not mandatorily nucleation due to the thermal conditioning, because non-volatile residuals of one certain size can grow to different particle sizes in the ambient aerosol. This would result in higher maximum concentrations in the non-volatile mode but not in an increased total number concentration.

2 Specific comments

1. "I suggest adding..."
   Both references were added. One sentence in the 2nd last paragraph of the introduction now refers to these early applications of aerosol volatility measurements:

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"Early applications of the thermodenuder in atmospheric studies involved the separation of the volatile and non-volatile fractions of accumulation and Aitken mode aerosol (Clarke, 1991; Smith and O'Dowd., 1996)."

2. "Figures 5-7..."
   The columns and rows were rearranged according to the reviewer's suggestion.

3. "Please specify particle number segment..."
   A particle number segment is a closed area under the particle number size distribution curve, delimited by an upper and lower diameter. For clarity, the 3^{rd} sentence of the 1^{st} paragraph in Section 3.3 was rewritten: "Briefly, the approach of the summation method is to associate segments of equal particle number concentration under both, the conditioned and unconditioned particle size distributions."

4. "Please comment on your observation stated in the abstract..."
   Two statements can be made here:

   (a) The Volatility Tandem DMA measurements performed at Melpitz (extracts shown in this paper; more technical details in Rose et al., 2006) showed a conservation of particle number during passage through a thermodenuder down to ambient diameters of 30 nm. Below that diameter, information could not be retrieved with confidence by the V-TDMA for that system was not able to detect residual particles smaller than 15 nm after the second DMA.

   (b) The use of TD-TDMPS and TDMPS size distributions allowed an examination of nucleation mode particles in the thermodenuder at even lower particle sizes, i.e., down to 3 nm. Figure 2 in Wehner et al. (2005) shows the time series of nucleation mode diameters that features parts of this data set in detail. In the Figure it can be seen that the non-volatile residues of the nucleation mode diameter could be tracked down to corresponding ambient sizes

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of 6–8 nm. Simultaneously, total particle number was found to be conserved within the measurement accuracy. In conclusion, even particles < 20 nm were found to have a non-volatile residue. Speculations have been made whether these non-volatile residues are of organic nature (e.g., polymers), but this cannot be verified using the techniques used here. The 5^{th} sentence of the abstract has been rewritten: "Within the measurement uncertainty, every ambient particle down to particle sizes of 10 nm is concluded to contain a non-volatile core in the observed continental background air."

References

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[Clarke(1991)] Clarke, A. D.: A thermooptic technique for in-situ analysis of size-resolved aerosol physicochemistry, Atmospheric Environment, 25A, 635–644, 1989.

[Rose et al.(2006)] Rose, D., Wehner, B., Ketzel, M., Engler, C., Voigtländer, J., Tuch, T., and Wiedensohler, A.: Atmospheric number size distributions of soot particles and estimation of emission factors, Atmos. Chem. Phys., 6, 1021–1031, 2006.

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