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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Spatio-temporal variability and principal components of the particle number size distribution in an urban atmosphere" *by* F. Costabile et al.

F. Costabile et al.

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We thank the referee for the remarks. Here are our answers to his questions.

♢ General remarks

• Question: p. 18164, l. 18: You state that measurement results of the different aerosol spectrometers should be comparable within 10 % in term of total number concentration. But did you also correct for deviations in size channels between the different instruments (besides the correction for diffusion losses as stated in the paper)?

○ Answer:

Yes, we corrected for deviations in size channels between the different instruments in

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that they were normalised to dry (i.e. RH < 5 %) conditions. As a first test during the evaluation of comparison measurements at the site S5, we checked whether the sizing of a size spectrometer was correct in comparison with the central TDMPS at S5. Deviations with respect to particle sizing were calculated preferentially for episodes when the size distributions of the ambient aerosol showed a significant peak, so that any mis-sizing could be detected with confidence. A comparison of all size spectrometers at the site S5 showed positive deviations between 0 and 6 % in terms of particle sizing. This overall span was not surprising, since some size spectrometers measure the aerosol with a recirculated sheath air at up to 30 % relative humidity while others, including the TDMPS at S5 measured dry aerosol at RH below 5 %Having determined, however, the shift in particle sizing for each instrument (corresponding essentially to a shift in modal peaks occurring in the size range between 30 and 100 nm), the corresponding size distribution diameters were scaled back by this factor, i.e. generating size distribution data roughly corresponding to "dry" conditions (i.e. < 5 % RH). We are aware of the limited accuracy of this procedure, for instance because the hygroscopic particle growth factor cannot be considered constant for all particle sizes, but think that our method works well enough to make the size distributions accurate in the region where the particle number maximum usually occurs. Based on these data, the number concentration efficiency was determined with respect to the instrument at S7, as described in our text.

• Question: Section 4.1: You give an overview of different case studies concerning homogeneous and heterogeneous aerosol events. Did you perform a sort of frequency study to have an estimate how often those cases appear during your study period/within a year?

○ Answer:

In view of the limited extension in time of the intensive spatial experiment and the limited extension in space of the long-term experiment, we thought our data-set to be not a representative statistical sample for the analysis of the occurrence frequency

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of the different case-studies. With this in mind, we did not implement any scientific protocol a day can be classified according to. In fact, the aim of our case-studies was to provide a basic phenomenology of the spatio-temporal variability of the aerosol particle size distribution, spanning from highly spatial homogeneous to totally heterogeneous aerosol events. On the other hand, in consideration of the remark of the referee, we have roughly estimated the frequency with whom the different case-studies occurred — by considering at least contemporary measurements at 4–5 sites. The results are reported in the following tables, showing the occurrence and frequency of occurrence of the case studies, respectively:

	C1	C2	C3	C4	C5	C6	Days
March	1	4	1	4	0	4	14
April	2	7	2	8	6	5	30
May	2	11	5	10	1	1	30
Total	5	22	8	22	7	10	74

	C1	C2	C3	C4	C5	C6
March	7.1 (%)	28.6 (%)	7.1 (%)	28.6 (%)	0.0 (%)	28.6 (%)
April	6.7 (%)	23.3 (%)	6.7 (%)	26.7 (%)	20.0 (%)	16.7 (%)
May	6.7 (%)	36.7 (%)	16.7 (%)	33.3 (%)	3.3 (%)	3.3 (%)
Total	6.8 (%)	29.7 (%)	10.8 (%)	29.7 (%)	9.5 (%)	13.5 (%)

C1. High spatial homogeneity of aerosols under sunny conditions
C2. High spatial homogeneity of aerosols under cloudy conditions
C3. A week-day with secondary new particle formation
C4. A week-day with limited pollution impact
C5. A week-day with high pollution impact
C6. A week-day featuring several aerosol types

A general observation is that, during a typical spring time in Leipzig, the aerosol particle distribution was highly homogeneous in space for 50 % of the days — cases C1, C2 and C3 —, and moderately homogeneous to inhomogeneous for the remaining 50 % — cases C4, C5, and C6.

♢ Specific remarks

• Question: p. 18158, l. 8: I think it should read 8216;more sensitive to..8217; instead 8216;more sensitive on..8216;

○ Answer:

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We agree.

• Question: p. 18177, l. 13: You calculated 8216;signature size distributions8217; from PCA coefficients (Fig. 7). I can not fully follow this approach. Could you please briefly comment on how those size distributions were calculated.

○ Answer:

We agree with the referee in that this point was probably not clear enough. In fact, in consideration of the length of the manuscript, we could not provide full detailed descriptions of our calculations. In figure 7, the "Signature size distributions" are represented by the un–standardised/normalised coefficients extracted by the Principal Component Analysis calculated in terms of the correlation matrix. We decided to calculate the Principal Component Analysis in terms of the correlation matrix in order the coefficients ($\alpha_{k,j}$) to be directly linked to — and to represent — the correlations between the original variables, i.e. the aerosol particle concentrations, and the PCs, i.e. the particular aerosol populations. Therefore, the coefficients would represent the relative weight of the aerosol particle concentrations in each particular aerosol population. We interpreted this as signature size distributions.

To compare more easily the values of the coefficients to the aerosol particle concentrations, we reported in figure 7 the coefficients $\alpha_{k,j}$ after their un–standardisation with respect to mean and standard deviation ($\alpha_{k,j} \cdot \sigma + \mu$), and after their un–log–normalization ($10^{\alpha_{k,j} \cdot \sigma + \mu}$). (This is the inverse procedure of data normalization and standardization according to we calculated the loadings reported in figure 5.) One might note that also the loadings ($\frac{\alpha_{k,j}}{\sqrt{\lambda_k}}$), reported in figure 5, have a meaning similar to the coefficients. However, the loadings are scaled by the amount of variance (λ_k) explained by each PC. They can, thus, be misleading in representing the particle size distributions.

• Question: p. 18178, l. 25: In the urban meteorology community the term 8216;urban canopy layer8217; is more established instead of 8216;roughness layer8217;. The term urban roughness (sub)layer refers to a certain layer (where fluxes are not constant

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with height due to the presence of roughness elements) in terms of turbulence in urban boundary layers.

○ Answer:

We agree with the remark.

• Question: p. 18184, l. 13: I can not follow your argument that traffic emission enhanced rural nucleation. Do you refer to advection of particles to the rural area downwind of Leipzig? Please briefly comment on this.

○ Answer:

Our findings (fig.6) suggest that, during 2 years, the particular aerosol population characterised by a clear nucleation mode at the rural site (PC4) was statistically more frequent and more internally correlated — i.e. had higher scores — during the working days than during the week-ends. An explanation for this statistical evidence might be given by a possible involvement of traffic emissions, which have typically weekly cycles in an urban environment. We have no evidence of where these traffic emissions came from - either local traffic emissions at the rural site (Melpitz) or traffic emissions of gaseous precursors from the downtown Leipzig or others. We have, however, indications that traffic emissions were likely to have been involved in the nucleation mechanisms at the rural site as well as in the urban area.

• Question: p. 18208, Fig. 5 a: In the text you state (p. 18174, l.3) that PC4 and PC5 illustrate urban traffic by a wide Aitken mode. However, only one of the roadside stations is elevated in PC 4 and PC5. Why not both roadside stations are commonly elevated above the other stations in both PC8217;s?

○ Answer:

We do appreciate this referee remark. This point would have probably required a better explanation. In fact, several findings strongly indicate urban traffic to be represented by both PC4 and PC5 in fig.5a. First, the statistical analysis at single sites only cannot

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fully recognize this aerosol population — the particle size range of PC3 at the site 5 in figure 4 is different from the wider particle size range of both PC4 and PC5 in figure 5a. Second, this aerosol population is highly correlated with traffic flows (tab.3), is more evident during working days (fig.8) and at roadside sites (PC4 and PC5 in fig.5a), but is still relevant also at background sites (fig.5 and fig.7). We, therefore, attribute the difference individuated by the referee to a different siting of the two sampling inlets. Despite the fact that both the traffic sites (No.3 and 5) are equally representative of roadside conditions, some dissimilarity may be noted: the sample inlet height (10 and 6 meters, respectively, tab.1) and the orientation of the two sample inlets with respect to the wind direction. These two differences could have induced two types of inhomegeneities in the results of the statistical analysis, which probably resulted in two different principal components at the two traffic sites with the same meaning. The different sample inlet heights could have resulted in different correlations between Aitken and nucleation mode particles — the difference between PC4 and PC5 in fig.5a is in the nucleation mode loadings, probably due to a different variability with height of the nucleation mode with respect to the Aitken mode. The different sample inlet orientations in the street-canyons could have generated a temporal shift between the two measurements at the two traffic sites - when the sample inlets were under different conditions in the street-canyon (i.e., downwind/leeward) with respect to both local and urban traffic emissions.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 18155, 2008.