Atmos. Chem. Phys. Discuss., 12, C12530–C12537, 2013
www.atmos-chem-phys-discuss.net/12/C12530/2013/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Temperature-dependent accumulation mode particle and cloud nuclei concentrations from biogenic sources during WACS 2010" *by* L. Ahlm et al.

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Received and published: 12 February 2013

We thank reviewer #1 for comments and suggestions for improvement of our manuscript. The comments from the reviewer followed by our responses to the comments can be seen below. In the updated manuscript, changes have been marked in red color.

Comment #1.

My only methodological problem is the use of correlations. First of all, they assume linear behaviour. This is neatly shown in Figure 10 for NCCN and T, but do such behaviour exist for other properties as well? There is no need for extra figures, but a

comment for using linear correlation could be useful. Related to this, fig 10a-b could be also (noisy) exponential, but I agree that there is too little data to make such conclusion. Also, use of coefficient of determination ("r2") could be more consistent with other literature. The main problem with correlations are, that only R value gives quite low degree of understanding on the processes included. The number of data points is not clearly defined (although could be deducted from the campaign length), and there is no indication of the level of autocorrelation (autocorrelation: corr(x(t),x(t+dt))) between the measurement points. The autocorrelation is significant, as can strongly affect how important different correlations are. The text uses now very qualitative comments on the correlation significance, which is not necessarily a bad thing, just leaves the reader to think how these are derived. For quantitative understanding of how "significant" * the results are could be determined by calculating some kind of confidence interval for the correlations given. This is needed in this article, as some of the conclusions are based on lower and higher correlations between different properties. Without any analysis, a R=0.3 and R=0.5 might actually be both completely insignificant (or both highly significant) and thus their difference might be either insignificant or significant. A word of warning: it is quite important to understand the role of autocorrelation to the correlation, as two strongly autocorrelating timeseries can easily give larger correlations between each other than non-autocorrelating ones. In (perhaps) more clear terms: If one calculates a classical t-test p-value for a correlation, the p value will most likely be strongly underestimated if the two timeseries are autocorrelating. For a methodology to do this, I would refer to some time series analysis book. I have used M. Mudelsee, Climate Time Series Analysis, Springer 2010 (ISBN 978-90-481-9481-0) and there algorithm 7.1. However, I leave the details (or even inclusion of this analysis) to the authors to include as they will. The key point, which I think will improve the level of discussion in this paper is to get some sort of quantitative level of understanding CAN one say anything about the differences in correlations. Please note that traditional p-values given most correlation coefficient routines (e.g. Matlab corrcoef) do not directly give reasonable values due high autocorrelation of datapoints.

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Response:

We have calculated 90% confidence intervals for all r-coefficients in Tables 1a-b, and Table 5. To calculate these confidence intervals, we first calculated the first-order autocorrelation for each time series, and used these to determine the effective data sizes that were finally used in the estimates of the confidence intervals. We think that a linear assumption in Fig. 10 is fairly justified by the plots. Perhaps we could get an even higher correlation if an exponential dependence was used in Fig. 10a-b, as the reviewer points out. However, we think that the correlation coefficients are high enough with the linear assumption to show that there is a strong dependence on temperature at both sites. When it comes to the linear assumptions between PMF factors and tracers (Table 1a-b), we think that a linear assumption makes most sense because both PMF factors and tracers are mass-based. At least we do not have enough information to assume any other specific relation. We have also added clearer information on how many data points were used for the correlation calculations.

Comment #2

pg 27994 - I assume LT has daylight savings time included? - When did the sun rise in the site? Approximate time is enough. This has some implications on understanding fig 8 and associated discussion.

Response:

Yes, local times include daylight savings. Sunrise occurred around 05:00 in the morning. This information has been added to Sect. 3.4.

Comment #3

Pg. 27997 - The authors refer to Macdonald et al ACPD paper, which is at least in the references still in preparation. I would like to ask them then to include slightly more information on the periods chosen. Eg. cooler, warmer, etc should be more detailed on what was the actual temperature range. One way would be to somehow include

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temperatures and precipitation to fig2, altough it seems to be rather full as it is.

Response:

We have added temperature and relative humidity for both sites to Fig. 2, and now refer to this plot in the text.

Comment #4

Pg 27999 Please detail MKV/MACR

Response:

MVK stands for methyl vinyl ketone, and MACR stands for methacrolein. The PTR-MS cannot resolve one from the other. Thus MVK/MACR represents the sum of the two compounds. We have added this information to the manuscript.

Comment #5

Pg 28005 Sect 3.4, line 23 on. The authors discuss some of the peaks in concentration and discuss their similarities and differences. This corresponds to period 3, and I would like to know more in detail how much of the speculated anthropogenic influences could be seen from the trajectory analysis as well. Additional comment is that the authors very carefully analyze each minor peak of the time series. Considering the usual variability of any atmospheric property, especially over a mountain site, I would perhaps not overanalyze each detail.

Response:

We calculated the potential source contribution function (PSCF) (e.g. Heo et al., 2009) based on the trajectories in Fig. 1 for the entire campaign. The attached file shows the PSCF plotted for the PMF combustion factor with a threshold value chosen as 0.5 μg m-3. Clearly, trajectories from southwest are associated with higher concentrations of the PMF combustion factor. For the trajectories from the north (Fig. 1 in the manuscript) where there are few anthropogenic sources, very few trajectories are as-

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sociated with combustion factor concentrations above the chosen threshold value. We made the same calculations for SO2 and for NOx but these plots looked more noisy, likely because there are local anthropogenic sources (which are less dependent on back trajectories) mixed with transport from Vancouver, Seattle. For this reason we did not find this discussion and these plots interesting and clear enough to add to the manuscript. The back trajectories in Fig. 1c also indicate westerly transport during period 3 from a sulphur source (smelter) located at the coast, suggesting a regional SO2 influence as well.

Comment #6

Pg 28008 When you say that combustion and N values correlate, which values do you use for N? Averages of the same periods?

Response:

We averaged these parameters over 24 hours and calculated the correlations for these averages. We have added this information to Table 4.

Comment #7

Pg 28009 I think the correlation between T and CCN is very interesting indeed. However, could this be happening from e.g. similar advection patterns? Could we get similar behaviour just from observing updrafts from valley below, which would typically have higher T as well as higher NCCN? Also, higher temperature could indicate higher updraft, and thus more influence from below. Please discuss.

Response:

The correlations are calculated for 24 hour averages of the parameters, and therefore do not include potential correlations due to diurnal variations resulting from e.g. anabatic and katabatic flows. This is mentioned in the figure caption of Fig. 10. There is of course still the possibility that days with higher temperatures had stronger updraft winds from the valley in daytime, and thereby possibly more transport of accumulation

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mode particles from the valley up to the mountain sites, which could result in higher average concentrations over 24 hours. However, if strong daytime anabatic flows would be the main cause for the strong correlations in Fig. 10, one would expect strong diurnal variations in N100-700 since katabatic flows dominate at night time, which should result in reduced concentrations. By looking at Fig 9c one can see that the diurnal variations in N100-700 are not very strong; at least they are small compared to lower-frequency variations in Fig. 9c. Hence, from this it seems that the prevalence of anabatic flows is not the reason for the correlations in Fig. 10. We have added some short discussion on this issue to the manuscript.

Comment #8

Fig 4: It might be good to include also your chosen labels for the factors in this figure (detritus, etc)

Response:

We have added these labels to Fig. 4.

Comment #9

Fig 9.a), sometimes it might be better to plot the aerosol concentrations in log-scale, to avoid the need of quite non-linear colour scale. However, the figure is perfectly readable as is. Authors mention earlier that some indication of biomass burning was evident in last days of the measurements. Are they seen in this figure somehow?

Response:

We tested to plot in log-scale but we found that the plot is clearest the way it is plotted now. The influence of biomass burning during the last days of the campaign is discussed in the overview paper, which is just about to be submitted. Since we prefer if these two papers do not overlap too much, we think it is best if biomass burning tracer data like e.g. acetonitrile, which provides a clear indication of biomass burning events, is presented there.

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References

Heo, J. –B., Hopke, P. K., and Yi S. –M.: Source apportionment of PM2.5 in Seoul, Korea, Atmos. Chem. Phys., 9, 4957-4971, 2009.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 27989, 2012.

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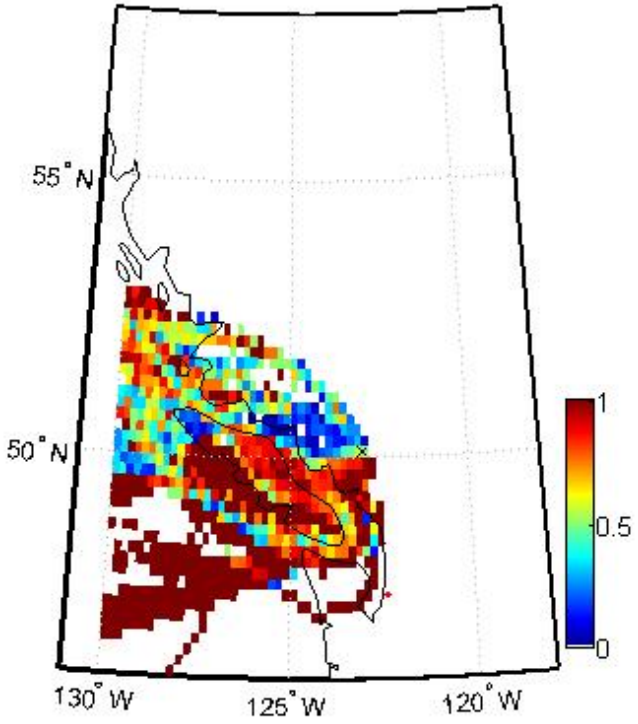

Fig. 1. PSCF for combustion factor (comment #5)

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