Atmos. Meas. Tech. Discuss., 3, C2720–C2726, 2011
www.atmos-meas-tech-discuss.net/3/C2720/2011/

Atmospheric Measurement Techniques Discussions

Interactive comment on "A two year's source apportionment study of wood burning and traffic aerosols for urban and rural sites in Switzerland" *by* H. Herich et al.

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Received and published: 16 February 2011

We thank the referee for the comments and will consider them in the revised version of the manuscript.

Please note: In the revised manuscript, the BC data from the three sites was extended until October 2010. The longer time series provide improved information about the temporal variation of considered measures (e.g. Angstrom exponent α) and even more robust results. In the revised version, the figures and the numbers in

tables have thus slightly changed. This has, however, no impact on the results and the conclusions.

This manuscript follows two existing papers in the application of online optical techniques for source apportionment analysis of biomass burning aerosols. The methodology applied is slightly modified with respect to the previous works, and the results are positive and presented in a clear and straightforward manner. As a result, the authors are able to suggest this methodology as a valid tool for source apportionment of carbonaceous aerosols. The paper is of interest and within the scope of AMT, and merits publication after revision.

Page 3, line 11: this does not apply to Southern EU, as biomass burning is not a major source of PM.

-We will change the text accordingly.

Please rephrase. Page 5, line 25: please provide some further details on the methodology, in case the readers have no immediate access to the paper referenced here. What is the methodology based on?

- We will very briefly describe the methodology in section 2.1, the following text will be included:

The authors applied a constant factor to correct for multiple scattering at the filter fibre in a unloaded filter as determined from studies with pure soot particles. To correct for increasing light attenuation due to accumulating particles in the filter (shadowing effect) an empirical function is used.

page 6, line 18: please specify, was it total K or soluble K? Only soluble K is a tracer of biomass burning, not total K

- It is water soluble K, we will clarify this in the text

page 6, lines17-22: the methodologies for the analysis of K and levoglucosan should be described.

- The following descriptions will be given in the revised manuscript:

The concentration of water soluble potassium was determined by ion-chromatography (Dionex IC 3000) after extraction of punches (2.5 cm diameter) of the daily PM2.5 (PAY) and PM10 filter samples (ZUE) in 40ml of nanopure water during approx. 15 hours.

Levoglucosan concentrations at PAY were determined by NILU as part of the intensive measurement periods of the European Monitoring and Evaluation Programme (EMEP) in fall 2008 and spring 2009. The applied method is described in Dye and Yttri (2005). Dye, C. and Yttri, K. E.: Determination of monosaccharide anhydrides in atmospheric aerosols by use of high-resolution mass spectrometry combined with high performance liquid chromatography, Anal. Chem., 77, 1853–1858, 2005.

Figure 2:the OC/EC ratio is not included in the legend page 8, line 25: please add “in summer” for the PAY data.

- Text will be changed.

Page 8 line 10 to page 9 line 5: an interpretation of the monthly and daily cycles of the absorption exponent would be welcome, in addition to the factual description of the variations currently provided by the authors.

- We added a general statement on page 8. The interpretation of the Angstrom exponent values is given in detail on page 9 lines 6-27.

The same applies to the description of the OC/EC ratios in Figs 2a and 2b. Page 10, line 8: regarding the selection of the absorption coeff of 0.9 for FF, what uncertainty do the authors associate to this value?

- Please see answer to Referee 1 on this point.

The selection of this 0.9 coefficient in this work seems very similar to the selection of the minimum OC/EC ratio representing primary vehicular emissions in the EC-tracer method. Despite the fact that in the EC-tracer method the minimum value is meant to be selected, it is currently under discussion whether that minimum value really represents primary vehicular emissions, and it has even been proposed that a factor of 0.5 should be applied to the minimum OC/EC value to obtain the real marker of primary vehicular traffic emissions. In the present work, what uncertainty do the authors consider? Could a similar debate originate as with the EC-tracer method? What is the range of values that the authors considered before selecting the 0.9 coefficient for FF, and how would different values have affected the final result? A sensitivity analysis would be very welcome here, or at least a discussion on the uncertainty.

- Again, please see the answer to Referee 1 for a discussion of the range of considered values for on α_{WB} and α_{FF} and for discussion of expected uncertainties. We don’t expect that a similar debate than for the EC-tracer method will originate. In literature, there is a consensus that diesel soot has an Angstom exponent close to 1 For the EC-tracer method, the ratio of primary EC to primary OC is estimated from ambient measurements, and the debate is whether this is possible or not. Here we rely on α_{WB} and α_{FF} as determined from emission measurements. There is certainly room for discussion about the values used however, we are confident that we covered

the range of reasonable values in the presented work.

Page 10, line 17: the assumption made by the authors that sources other than biomass burning and fossil fuels affect the carbonaceous matter mass is rather evident. However, the model seems to have worked in previous papers (Sandradewi et al and Favez et al). How do the authors explain this? Why should the length of the study period have an impact on this, as suggested by the authors?

- Sandradewi et al 2008a and Favez et al 2009 use only winter data. In winter the impact of SOA might be negligible and the "two sources approach" can be appropriate. However, when using data covering the whole year, we see that the "two sources approach" is not capable to explain the variability of carbonaceous matter. The model is incomplete probably due to the impact of significant fractions of carbonaceous matter from other sources or processes than biomass burning and fossil fuel combustion (e.g. SOA). Also when we used the "improved Aethalometer model" (including an intercept in Equation 4) as proposed by Sandradewi et al 2008b and Favez et al 2010 the method did not lead to a satisfactory model for our data. We discuss this in detail in p.10-p.11.

Figure 3: why is the correlation between measured EC and modelled BC so low in ZUE, and especially much lower than at PAY and MAG? Page 12, line 18: I find the total BC concentrations in ZUE rather low (0.99 to 1.34), do these results agree with previous studies and/or other works?

-For ZUE, EC data for comparison with the aerosol absorption measurements with the aethalometer are currently only available for the time from Apr.09 to Dec.09. No data is currently available for January to March, when air pollutant concentrations (such as EC) are typically highest due to meteorological conditions favouring accu-

mulation of air pollutants in the boundary layer. This and the fact that the number of data pairs in Figure 3 is for ZUE lower than at the other sites might explain the lower observed correlation between aerosol absorption and EC. However, it seems that in ZUE the number of outlying data points is relatively high compared to the other sites leading to a relatively high uncertainty for the aerosol light absorption cross section at ZUE (see Table 2).

Page 12, lines 9-20: the mean contribution of BCwb in wither is very similar for all three sites (29%, 27%, 24%). Was this result expected? Wouldn't WB emissions be much higher in the rural areas with respect to ZUE?

- With the extended data, the relative contributions of WB in winter changed a bit, now ranging from 24% in ZUE to 33% in PAY (30% in MAG). However, the order of sites changes when absolute contributions of BC_{WB} to total BC are considered, because BC levels are highest at MAG followed by ZUE and PAY. This is not surprising when taking a closer look at the characteristics of the considered measurement sites: In contrast to MAG, PAY is not influenced by emissions from wood combustion appliances located in the near vicinity of the site but represent more the rural background situation in that region. On the other hand it is known from other studies (cited in the manuscript) that wood combustion has significant contributions to the levels of PM in urban ZUE. As indicated in the manuscript, the results for ZUE are in good agreement to the findings of Szidat et al. 2005, for MAG and PAY no similar studies are available.

Figures 5 and 6: the data are not very visual in the current scatter plots; the authors could maybe consider other ways of representing the data? Or simply separating the FF and WB correlations in 2 different plots, that might already help.

- We tried a few things but did not find a better solution for representing the dependence between estimated contributions of BC_{WB} and BC_{FF} and other indicators for WB emissions. We feel that Figures 5 and 6 are appropriate for getting the information and suggest to keep them as they are.

Interactive comment on Atmos. Meas. Tech. Discuss., 3, 5313, 2010.