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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Contribution of residential wood combustion to hourly winter aerosol in Northern Sweden determined by positive matrix factorization" *by* P. Krecl et al.

P. Krecl et al.

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We thank anonymous referee #2 for the constructive and helpful criticism. A detailed response to the comments of reviewer #2 follows below.

Specific comments

1. Title of paper. The title has been changed to better describe the content of the manuscript as suggested by the reviewer: “Contribution of residential wood combustion and other sources to hourly winter aerosol in Northern Sweden determined by positive matrix factorization”.

2. Page 5730; 36; shorten PMF method. We agree with the referee that the PMF

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method has been extensively characterized in the literature. However, the application of PMF to particle number size distributions is quite novel. We think that shortening the description of the method applied to particle number size distributions and definition of several variables and parameters we use later in the article would make our presentation less clear to the general reader.

3. Methodology: Multiple linear regression. Since multiple linear regression is a widely used statistical method we only included a short text as suggested by the referee. “Following Hedberg et al. (2006), a multiple linear regression model of the g-factors onto the measured concentrations of each aerosol variable (i.e. N25-606, MLAC, PM10, and PM1) was performed. The MLR model assumes that the concentrations of each aerosol variable can be expressed as a linear function of the g-factors and determines the regression coefficients and their confidence intervals (95%). The source contributions to each aerosol variable in then estimated based on the calculated regressions coefficients.”

4. Page 5737, line 9. Does MLAC originate only from diesel cars? Any references? The origin of the factor 1 is most likely to be local vehicle exhaust emissions since the modelled particle number size distribution for this factor is similar to the shape of particle number size distributions measured at a street canyon and a road tunnel sites in Stockholm (with similar vehicle fleet as in Lycksele). The low percentage of diesel traffic in Sweden could explain the weak correlation between modelled N25-606 for factor 1 and the measured MLAC found in this study. Diesel vehicles produce a larger percentage of light-absorbing carbon aerosols compared to spark-ignition engines as found by Burtscher (2000) and Gillies and Gertler (2000). The sentence has been rewritten in the manuscript to clarify this point and include references: “The weak correlation ( ) we found between modelled N25-606 for factor 1 and measured MLAC could be explained by the dominant gasoline vehicle emissions since gasoline vehicles produce lower MLAC than diesel engines (e.g., Burtscher, 2000; Gillies and Gertler, 2000).”

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5. Page 5738, line 18. “Author suggests that factor 3 consist of two sources: local wood combustion and long-range transport. Can you see this from the size distribution of factor 3 (Fig. 4)? Can you assume that the size distribution of local RWC in factor 3 is similar to the size distribution of factor 2?” Several laboratory studies show that the particle number size distribution of wood combustion is highly dependent on the burning conditions and type of appliance (e.g., Hedberg et al., 2002; Johansson et al., 2004; Boman, 2005). These studies generally indicate that during less efficient combustion the number concentration of fine particles decreases and the particle size increases compared to more optimized combustion conditions. In the case of inefficient burning, the mode number size distribution is similar to the peak number size distribution of the Nordic background aerosol. Then the PMF model is not able to completely detach the contribution of RWC (inefficient burning conditions) from that of long-range transport. Then, the particle number size distributions of local RWC in factor 3 is different from the size distribution of factor 2. Factor 2 most likely corresponds to local RWC under more optimized burning conditions whereas factor 3 is a combination of local RWC (less efficient combustion) with long-range transport contribution.

The text has been rewritten as follows: “As discussed above, RWC can produce substantially different particle size distributions with different particle diameters at peak number concentrations. During less efficient combustion the number concentration of fine particles in the emission tend to decrease and the particle size increases compared to more optimized combustion conditions (Boman, 2005). In this study, number size distributions from local RWC (poorer combustion conditions) and LRT might partially overlap.”

6. Page 5741, lines 12&#8211, 16. “By comparing the fraction of local RWC obtained in this study to that of Hedberg et al. (2006) suggests that the fraction is closer to possible maximum than minimum. Does this mean that the size distribution for RWC is bimodal? Does this agree with other studies?” We do agree with the referee that according to the results of Hedberg et al. (2006) the contribution of

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local RWC to the aerosol mass concentration is closer to the maximum than to the minimum threshold we found in the Lycksele experiment in winter 2005/2006. The fact that the contribution of RWC to the aerosol mass concentration is dominant does not necessarily mean the particle size distribution should be bimodal. In our case, we measured particle number size distributions with a DMPS system in the submicron fraction. Assuming that the particles are spherical and have certain density, the particle mass size distribution in the 25-606 nm range could be estimated from the measured particle number size distributions. However, we do not have enough information to determine if the particle mass size distribution is bimodal when considering all particle diameters (i.e. not only the fine fraction). The comparison of our results with laboratory studies and other field studies has already been presented in Krecl et al. (2007b). A summary follows: As mentioned before, the operating conditions of the wood burning appliance have a strong impact on the particle size distribution of the emitted aerosol. Many laboratory studies found unimodal particle number size distributions for wood combustion (e.g., Nussmaumer, 2001; Boman, 2005) while Hedberg et al. (2002) reported bimodal particle number size distributions in the submicron range. Particle number size distributions measured in the field in areas impacted by RWC showed unimodal particle number size distributions. For example, Hering et al. (2007) found that the mean particle number size distribution for evening periods peaked at ˜80 nm in Fresno (USA) during the winter when RWC is very common in the area. A previous study in Lycksele in winter 2001/2002 by Kristensson (2005) also found unimodal particle number size distributions with the mode at ˜70 nm.

7. Page 5741, line 16&#8211, 23. “How can you compare fossil total organic carbon to MLAC? I assume that MLAC, measured using an aethalometer, consists mostly of inorganic (elemental) carbon.” We agree with the referee that the text was unclear and confusing. The term “total organic carbon” employed by Sheesley et al. (in preparation, 2008) is defined as the carbon remaining (EC+OC) after removal of inorganic carbonates by acid treatment. Then the carbon quantification is carried out by C/H/N elemental analysis. The Aethalometer measures

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the light-absorbing carbon fraction. To avoid confusions with the usual atmospheric aerosol terminology, we changed the notation and renamed the “total organic carbon” as total carbon. The text has been modified accordingly: “Finally, our apportionment of MLAC can be roughly compared to the radiocarbon analysis results of total carbon reported by Sheesley et al. (2008) when sampling in Lycksele from 23 January to 8 March 2006. Total carbon is defined as the carbon remaining after removal of the inorganic carbonates by acid treatment and, thus, includes elemental and organic carbon. If we assume that the fossil total carbon fraction absorbs light in the same amount as the modern total carbon fraction does, then the apportionment found with radiocarbon analysis might apply to the MLAC data. To correctly interpret and compare these results, the reader has to bear in mind that 14C analysis provides the apportionment of modern and fossil carbonaceous aerosol but does not provide information on the location of the emission sources (local or LRT). The average contribution of fossil total carbon (attributed to traffic emissions) was 24% which coincides with the mean fraction of MLAC we attributed to local traffic emissions. The other 76% was mostly attributed to wood combustion since biogenic emissions and combustion of grass fires and incineration of household vegetable waste were not observed in the area.”

Technical correction Page 5730, line 4 and reference list: “Taaper” was replaced by “Tapper” in both occasions.

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