

Interactive comment on “On the growth of nucleation mode particles: source rates of condensable vapor in polluted and clean environments” by M. Kulmala et al.

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The authors would like to thank the anonymous referee 2 for constructive comments.

General questions:

The referee is concerned of the misinterpretation of primary emissions as secondary aerosol production in urban environment. Although the concern is justified, the primary emissions can be detected based on the visual inspection of the evolution of observed size distributions. Our instruments measure the aerosol size distribution resulting from both primary sources as well as secondary aerosol production and growth. Since the primary particles are emitted as they are, they appear suddenly to the size distribution

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and they do not substantially grow during the day. The method used in this study relies on the growth rate calculations and is not affected by the primary particle production. However, as the referee points out, the observed growth rates are effected by growth due to coagulation. This is not explicitly taken into account. The effect is of minor importance in the rural and remote sites but in the polluted sites, New Delhi in particular, it can contribute and thus increase the uncertainty of the estimated source rates of condensable vapours. On the other hand, it cannot explain the order of magnitude differences in the source rates between the different sites.

The author response to the comment concerning the representativeness of short campaigns is taken into account and a chapter is added to the manuscript (Results and discussion page 6951 line 19 onwards)

"Taken into account the high seasonal variability of the ambient aerosol size distributions, the calculated condensation sinks and growth rates as well as estimated condensable vapour concentrations and source rates should be used with caution. This concerns particularly short field campaigns, in which the dataset is quite limited. The long-term measurements, on the other hand, can be used to represent characteristic values of the region over the proposed period (SMEAR I: all seasons, SMEAR II: winters)."

Specific comments:

Page 6947 lines 19-25: The size ranges presented in the paper are typical ranges indicating a variation and even overlap between the system specifications during different campaigns. The difference between the two systems was monitored during campaigns, and systems were intercompared in more detail, if an inconsistency was observed.

Page 6948 lines 24-30: The drying is explicitly mentioned in Athens and Marseille cases due to fact that during these campaigns, external drier was placed in the inlet, which ensured drying of particles but also increased sampling losses. During the other campaigns and long-term measurements, no external drying was commenced. The

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ambient aerosol size distributions were monitored as dried particles. This was ensured with RH-sensors placed in the sheath flow of the differential mobility analyser. The difference between the dried aerosol sample size distributions and prevailing ambient conditions were not taken into account in the analysis. This would need either modifications in the instruments or assumption of the hygroscopic growth of sampled particles with size and RH dependency.

Page 6949 lines 10-13: The absolute sink provided by the supermicron particles is truly much higher in the polluted urban site of New Delhi. The sentence is changed as follows: "However, the effect of larger particles to the condensable vapor concentrations and source rate estimations were typically less than 5 per cent in Hyytiälä and coastal Mace Head site in Ireland (Dal Maso et al., 2002), which justified the neglect of the sink provided the supermicron particles in the rural and remote locations (Hyytiälä, Värriö and Antarctica). In the polluted urban site of New Delhi the absolute value of the sink provided by supermicron particles could be significantly higher than at cleaner sites, but its contribution to the overall sink is likely to be smaller. For the calculated condensable vapors, however, the ratio between sink in the sub- and supermicron particles is the most important factor. Based on this argument, the uncertainty of the estimated condensable vapor concentration caused by neglecting supermicron particles is probably lower in the polluted environment than in cleaner regions."

Page 6949 Line 21: Inconsistency of the lengths of the campaigns is corrected as proposed by the referee.

Page 6955 lines 4-7: The whole chapter is reformulated: "The estimation of vapor source rates and concentrations using this quite straightforward technique agrees well with box model studies, which included detailed aerosol dynamics (Kulmala et al, 1998, 2000). So it seems that despite the simplifications in the calculations, the obtained estimations of the condensable vapors and its source rates is possible with the simple analytical method."

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Page 6960 Table 1: The discussion on uncertainties is enlarged in the results and discussion part of the paper (Page 6952, line 12 onwards: "Experimental parameters needed in the calculations of condensable vapor are determined with a maximum uncertainty of a factor of 2. This uncertainty cannot explain the magnitude differences in the calculated concentrations and source rates of condensable vapors.")

Page 6962 Figure 2: The sudden shift in the observed size distribution during Athens campaign can be explained by a slight change in local wind direction. The measurement site was located at the northern border of the city and during the afternoon hours the wind was turning to blow from the urban region.

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 6943, 2004.

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