Atmospheric Chemistry and Physics
An Interactive Open Access Journal of the European Geosciences Union

Atmos. Chem. Phys., 5, 515-532, 2005
www.atmos-chem-phys.net/5/515/2005/

To what extent can aerosol water explain the discrepancy between model calculated and gravimetric PM_{10} and $PM_{2.5}$?

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Abstract. Inter-comparisons of European air quality models show that regional transport models, including the EMEP (Co-operative Programme for monitoring and evaluation of the long-range transmission of air pollutants in Europe) aerosol model, tend to underestimate the observed concentrations of PM_{10} and $PM_{2.5}$. Obviously, an accurate representation of the individual aerosol constituents is a prerequisite for adequate calculation of PM concentrations. On the other hand, available measurements on the chemical characterization of ambient particles reveal that full chemical PM mass closure is rarely achieved. The fraction unaccounted for by chemical analysis can comprise as much as 30-40% of gravimetric PM_{10} or $PM_{2.5}$ mass. The unaccounted PM mass can partly be due to non-C atoms in organic aerosols and/or due to sampling and measurement artefacts. Moreover, a part of the unaccounted PM mass is likely to consist of water associated with particles. Thus, the gravimetrically measured particle mass does not necessarily represent dry PM_{10} and $PM_{2.5}$ mass. This is thought to be one of the reasons for models under-prediction of observed PM, if calculated dry PM_{10} and $PM_{2.5}$ concentrations are compared with measurements. The EMEP aerosol model has been used to study to what extent particle-bound water can explain the chemically unidentified PM mass in filter-based particle samples. Water content of $PM_{2.5}$ and PM_{10} has been estimated with the model for temperature 20°C and relative humidity 50%, which are conditions required for equilibration of dust-loaded filters according to the Reference method recommended by the European Committee for Standardization (CEN). Model calculations for Europe show that, depending on particle composition, particle-bound water constitutes 20-35% of the annual mean PM_{10} and $PM_{2.5}$ concentrations, which is consistent with existing experimental estimates. At two Austrian sites, in Vienna and Streithofen, where daily measurements of $PM_{2.5}$ mass and chemical composition are available, calculated $PM_{2.5}$ water content is found to be about 75-80% of the undetermined $PM_{2.5}$ mass and there is correlation between them. Furthermore, accounting for aerosol water has improved the agreement between modelled and measured daily $PM_{2.5}$ concentrations, whilst model calculated dry $PM_{2.5}$ concentrations appear to agree quite well with the total identified $PM_{2.5}$ mass. No information on the composition of PM measured at EMEP sites is presently available. Given that PM_{10} and $PM_{2.5}$ concentrations are measured at EMEP stations with gravimetric methods they are likely to contain water. We show that the levels of modelled PM_{10}

and $PM_{2.5}$ concentrations with aerosol water included agree with measurements better than dry PM concentrations. As expected, the spatial correlation has not changed significantly, whereas the temporal correlation of daily PM_{10} and $PM_{2.5}$ with monitoring data has slightly improved at most of the EMEP sites. Our results suggest that aerosol water should be accounted for in modelled PM_{10} and $PM_{2.5}$ when compared with filter-based gravimetric measurements.

Final Revised Paper (PDF, 2138 KB) Discussion Paper (ACPD)

Citation: Tsyro, S. G.: To what extent can aerosol water explain the discrepancy between model calculated and gravimetric PM_{10} and $PM_{2.5}$?, Atmos. Chem. Phys., 5, 515-532, 2005. Bibtex EndNote Reference Manager