FINAL PUBLISHABLE SUMMARY REPORT

Quantifying the effect of aerosols on the planet's radiative balance is one of the most urgent tasks in our efforts to better understand air quality and future climate change. The uncertainty in aerosol radiative forcing is large, which makes the evaluation of climate sensitivity difficult. The contributions of the various aerosol sources, the role of long-range transport, and the contribution of primary and secondary particulate matter to the ambient aerosol concentrations over Europe are still largely unknown. In this project we have made important steps towards a better understanding of the processes involved in aerosol chemistry and physics with a special focus on i) improved representation/prediction of aerosol number concentration in regional chemical transport models, ii) improved representation/prediction of aerosol mass concentration in regional chemical transport models, iii) organic aerosol, the least understood component of ambient particulate matter, and iv) identifying potentially efficient emission control strategies for Europe.

A three-dimensional regional chemical transport model (CTM) with detailed aerosol microphysics, PMCAMx-UF, was applied to the European domain to simulate the contribution of direct emissions and secondary formation to total particle number concentrations. PMCAMx-UF uses the Dynamic Model for Aerosol Nucleation and the Two-Moment Aerosol Sectional algorithm to track both aerosol number and mass concentration using a sectional approach. The model predicts nucleation events that occur over scales of hundreds up to thousands of kilometers especially over the Balkans and Southeast Europe. The model predictions were compared 10 against measurements from 7 sites across Europe. The model reproduces more than 70% of the hourly concentrations of particles larger than 10nm within a factor of 2. About half of these particles are predicted to originate from nucleation in the lower troposphere. Regional nucleation is predicted to increase the total particle number concentration by approximately a factor of 3. For particles larger than 100nm the effect varies from an increase of 20% in the eastern Mediterranean to a decrease of 20% in southern Spain and Portugal resulting in a small average increase of around 1% over the whole domain. Nucleation has a significant effect in the predicted N50 levels (up to a factor of 2 increase) mainly in areas where there are condensable vapors to grow the particles to larger sizes. A semi-empirical ternary sulfuric acid-ammonia-water parameterization performs better than the activation or the kinetic parameterizations in reproducing the observations. Reducing emissions of ammonia and sulfur dioxide affects certain parts of the number size distribution.

In this project we also examined the role of horizontal grid resolution on the performance of the regional three dimensional CTM PMCAMx. Two cases were investigated. First, the model was applied over the North Eastern United States with grid resolutions of 36 and 12 km during both a summer and a winter period. In this case the emission inventory was simply interpolated from the low resolution version. In the second case a multi-scale 36/4 km grid resolution is used over Western Europe

with high resolution (1 × 1 km) emissions. The improvement in model predictions with the fine grid is modest during the summer and more significant during wintertime at both domains. Major differences are predicted mostly for primary (organics and black carbon) rather than secondary (e.g. sulfate) species. The use of high grid resolution decreases the bias for black carbon concentration by more than 30% in the North Eastern US during wintertime. In the Megacity of Paris, using high resolution emissions and a 4-km grid decreases the fractional bias for organic aerosol from 80% to 60% in the city center and produces much larger spatial concentration gradients in the domain as compared to the emissions being interpolated. These results suggest that the grid resolution could improve model predictions; however, it is not currently the major source of the discrepancies between model predictions and observations.

Multigenerational oxidation chemistry of atmospheric organic compounds and its effects on aerosol loadings and chemical composition was also investigated by implementing the Two-Dimensional Volatility Basis Set (2-D-VBS) in a Lagrangian host chemical transport model. Three model formulations were chosen to explore the complex interactions between functionalization and fragmentation processes during gas-phase oxidation of organic compounds by the hydroxyl radical. The model was applied to summer and winter periods at three sites where observations of organic aerosol (OA) mass and O:C were obtained all over Europe. The base case model reproduces observed mass concentrations and O:C well, with fractional errors (FE) lower than 55 % and 25 %, respectively. The detailed functionalization scheme tends to overpredict OA concentrations, especially in the summertime, and also underpredicts O:C by approximately a factor of 2. The detailed functionalization model with fragmentation agrees well with the observations for OA concentration, but still underpredicts O:C. Both heterogeneous oxidation and aqueous-phase processing have small effects on OA levels but heterogeneous oxidation, as implemented here, does enhance O:C by about 0.1. The different schemes result in very different fractional attribution for OA between anthropogenic and biogenic sources.

PMCAMx-2008 was also applied in Europe to quantify the changes in fine particle ($PM_{2.5}$) concentration in response to different emission reductions as well as to temperature increase. A summer and a winter simulation period were used, to investigate the seasonal dependence of the $PM_{2.5}$ response to 50% reductions of SO_2, NH_3, NO_x, anthropogenic VOCs and anthropogenic OA emissions and also to temperature increases of 2.5 and 5 K. The substantial reduction of $PM_{2.5}$ components due to emissions reductions of their precursors in conjunction with significant changes of PM after increasing the temperature indicate that both emissions and temperature need to be of significant concern for improving air quality.