Geophysical Research Abstracts
Vol. 14, EGU2012-59, 2012
EGU General Assembly 2012

Simulating ultrafine particle formation in Europe using a regional CTM: Contribution of primary emissions versus secondary formation to aerosol number concentrations

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PMCAMx-UF, a 3-D regional chemical transport model with detailed aerosol microphysics, was applied to the European domain for the first time, in order to simulate particle number concentrations. Results for the month of May 2008 are presented during which an intensive measurement campaign was performed in Europe. The focus of this work was to better understand the relative contribution of direct emissions and new particle formation to regional aerosol number concentrations in Europe during a photochemically active period. Results show that nucleation events start generally from Eastern Europe and move to the west following the photochemical activity. The model predicts nucleation events that occur over scales of hundreds up to thousands of kilometers mainly in the high SO_2 areas of the Balkans and Southeast Europe as well as more localized events in west and central Europe. Aerosol nucleation is predicted to increase the total particle number concentration by a factor of 20 or more in some of these wide areas and by a factor of 6.5 for particles larger than 10 nm. The effect of nucleation in the lower troposphere on particle number above 100 nm is, on average, small with the exception of the Mediterranean area where the model predicts that nucleation can increase the number concentration of CCN-sized particles by up to 20%.

Model evaluation was conducted against ground based hourly measurements from 7 sites. The model agrees encouragingly well with the particle number concentration ground measurements, reproducing more than 70% of the (hourly) data points for N10 within a factor of 2. However, a systematic underprediction of N100 implies the need for improvement of either the size distribution of the emissions and/or the growth of the ultrafine particles to this size range. Approximately 50% of particles above 10 nm are predicted to come from aerosol nucleation. Including nucleation in the simulation has a minor effect in the predictions of N100 at all studied sites, ranging from -4% to +3% while the effect on N50 is more pronounced only in sites that are influenced by local pollution and have high aerosol number concentrations. Results from the 3 nucleation mechanisms that were tested suggest that i) the ternary mechanism performs better than the activation or the kinetic, and ii) additional improvements in the diurnal and geographical distribution of the particle number emission inventory are needed.