MODELLING OF PARTICULATE MATTER IN EMEP

1. Model calculation | 2. Verification | 3. Development

The impact on human health by atmospheric particles has been the recognized driving force for the extension of EMEP framework to include particulate matter. The preliminary assessment by WHO-EMEP (WHO-EMEP, 1999) and a number of more recent studies have indicated a significant association between the long range component of particulate matter, measured as the total mass of PM10, and a wide range of health damaging effects. Aerosols affect the radiative balance and thus contribute to climate change. Visibility impairment is another adverse effect of the atmospheric aerosol.

1. Model calculation of PM10in Europe

The first evaluation of the particulate matter mass in 1998 in Europe has been made based on separate estimates of the contributions from secondary inorganic aerosols (SIA), secondary organic (SOA) aerosols and primary particulate matter (PPM) (Tarrason et al., 2000). Concentrations of SIO, i.e. sulphate, nitrate and ammonium, have been computed with the EMEP Eulerian acid deposition model. The Largangian photooxidant model has been used to estimate 3-year (1993, 1995 and 1996) average for biogenic SOA, which were found to be the major contributor to the total SOA (Andersson-Sköld and Simpson, 2000). The atmospheric concentrations of primary PM10 has been calculated with a special version of the EMEP Eulerian dispersion model. The model uses the same as the EMEP Eulerian acid deposition model horizontal, vertical and temporal resolutions and common description of advection and diffusion. The model has been modified to give a better description to aerosols dry and wet deposition (Tsyro and Erdman, 2000). Emissions of primary PM10 and PM2.5 in 1990 have been derived from the TNO emission inventory (Berdowski et al., 1998).

The most recent EMEP assessment of PM10 mass concentrations, as the sum of SIA and PPM, have been made with the EMEP Eulerian models for the meteorological conditions of 1999 (Tsyro and Tarrason, 2001). For the calculations of SIA, officially submitted by Parties to the Convention emissions of SO2, NOx and NH3 in 1999 (Vestreng, 2001) have been used. The air concentrations of primary particles have beed calculated using updated emissions of PM2.5 and PM10 in 1995 compiled by the Netherlands Organisation for Applied Scientific Research (TNO). Due to rather large uncertainties involved in modelling secondary organic aerosols and given their smaller contribution to the total aerosol mass, SOA have not been included in the present calculations.

Estimated total mass concentration of PM10 in Europe is presented in Figure 1.

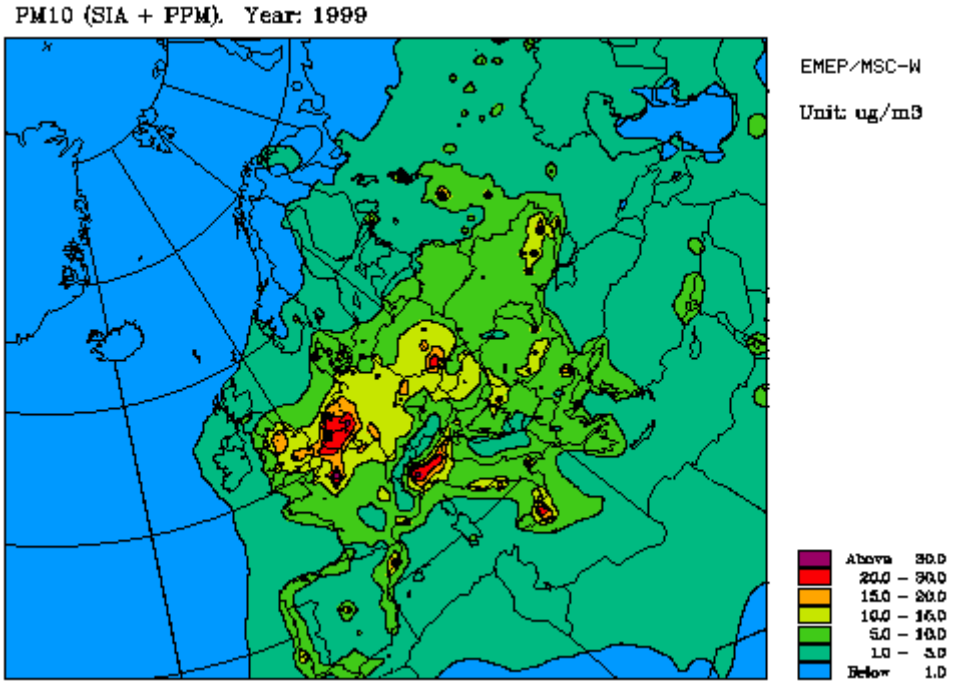

Figure 1. Annual mean concentration of total particulate matter in 1999. Unit: µg/m3

See also the figures for individually calculated annual mean concentrations of SIA (sulphate, nitrates, ammonium) in Figure 2 and primary PM10 in Figure 3.

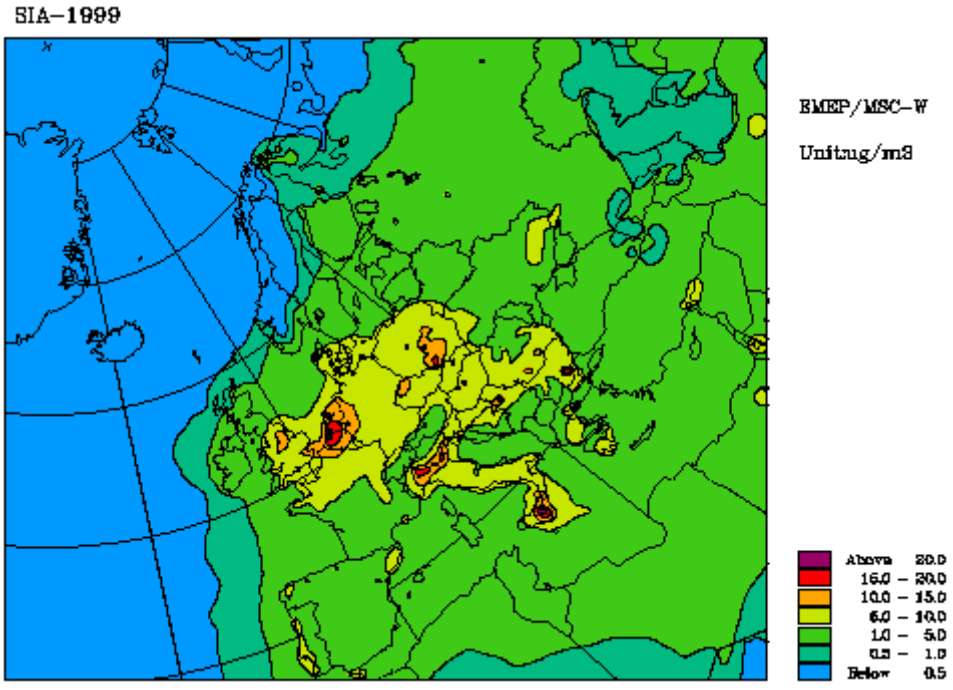

Figure 2. Annual mean concentration of SIA in 1999. Unit: µg/m3

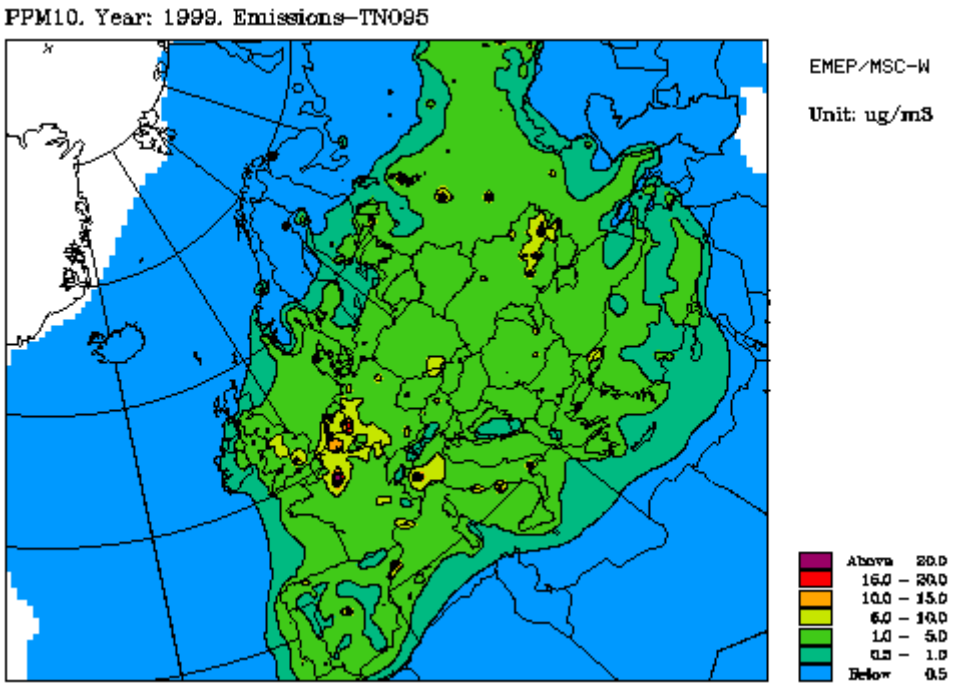

Figure 3. Annual mean concentration of primary PM10 in 1999. Unit: µg/m3

2.Verification of modelled PM10

Figure 4 compares the model calculated annual mean PM10 with concentrations of particulate matter in 1999 measured at EMEP stations. A general underestimation by the models of aerosol concentrations is partly because only SIA and primary particles have been included here, while such particles as re-suspended anthropogenic and natural mineral dust, sea salt, biogenic aerosols which can contribute considerably to PM mass have not been accounted for in the present calculations. Another reason for model underestimation of the total PM10 is that SIA concentrations are reported underestimated by the EMEP Eulerian acid deposition model. Considerable discrepancies between the calculated and measured values at Spanish and Italian stations are also because concentrations of total suspended particles (TSP) have been reported at those sites. In Spain, Saharan dust is expected to be rather a large contributor to PM mass. Further discussions on evaluation of the PM calculations, as well as the verification of modelled chemical composition of aerosols can be found in Tsyro and Tarrason, 2001.

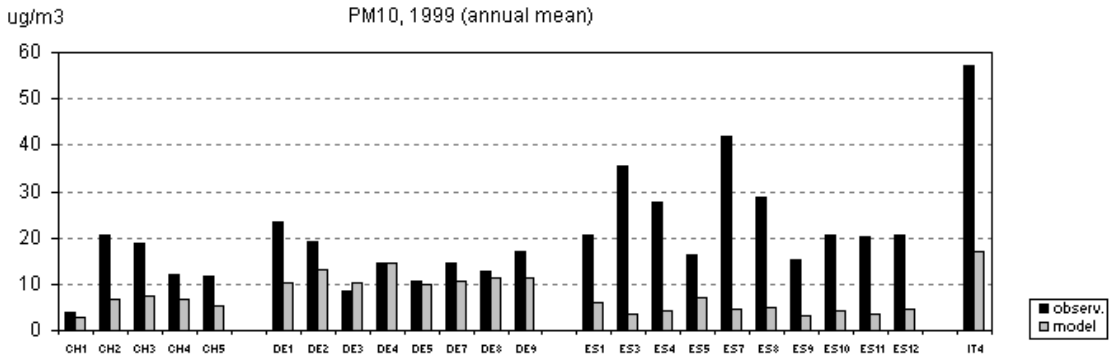

Figure 4. Model calculated and measured at EMEP stations annual mean concentrations of PM10 in 1999. Unit: µg/m3

3. Development and testing aerosol dynamics module

To provide necessary for policy development information on the aerosol number and size distribution and its chemical composition, an aerosol dynamics module is presently under implementation in the Unified EMEP Eulerian model. The aerosol model has been designed to respond to policy needs on the source-allocation of atmospheric particles and to facilitate the evaluation of aerosols adverse health effects. To this purpose, the model aims at calculating long-term information on the size and number distribution and chemical composition of particulate matter in Europe under EMEP.

MULTIMONO

multicomponent monodisperse aerosol dynamics model

(University of Helsinki, EMEP/MSC-W)

	N	SO4	NO3	NH4	EC	OC	dust	Sea salt	Water
Nucleation D< 0.02 µm	♣	♣							Diagnos-tic
Aitken 0.02 <D< 0.1 µm	♣	♣	♣	♣	♣	♣	♣		parameter
accumulation 0.01 <D< 2.5 µm	♣	♣	♣	♣	♣	♣	♣	♣	
coarse 2.5 <D< 10 µm	♣	♣	♣	♣	♣	♣	♣	♣	

Assumption: all particles within each mode have the same size and chemical composition, internal mixture radius: $R_i <= (\Sigma M_i, N_i)$

Processes: nucleation (ternary NH_3 - H_2SO_4 – H_2O), growth by condensation and coagulation

Figure 5.The aerosol dynamics module MULTIMONO.

The aerosol dynamics module MULTIMONO allows for nucleation, condensation and coagulation of particles and therefore is expected to give a better description to the aerosol atmospheric transport (Figure 5). The aerosol model will account for seven chemical components, namely, sulphates, nitrates, ammonium, elemental and organic carbon, sea salt and mineral dust, which are assumed to occur in an internal mixture. The size distribution of atmospheric particles is represented by four modes: nucleation (d<=0.02 µm), Aitken (0.02<d<=0.1 µm), accumulation (0.1<d<=2.5 µm) and coarse (2.5<d<=10.0 µm). The aerosol dynamics module is a multicomponent monodisperse model developed at the University of Helsinki (Pirjola and Kulmala, 2000) in a close co-operation with EMEP. The main advantage of the monodisperse approach is that it limits the number of prognostic variables to only two, namely number and mass, thus allowing for computationally efficient source allocation. Comparison of the monodisperse aerosol model with more sophisticated sectional model has shown that monodisperse approach provides an appropriate accuracy of description of the evolution of atmospheric aerosols, including processes of formation of new particles through nucleation, particles growth due to condensation of gaseous species and coagulation.

Prior to the implementation in the Eulerian dispersion model, MULTIMONO has been tested with a box-model with respect to the integration scheme and the effect of different parameters, e.g. initial particle concentrations, concentration of condensable gases and meteorology, on the aerosol dynamics processes. Figure 6 compares 24-hour evolution of particles number and size calculated with the sectional model AEROFOR and MULTIMONO. New particles form by ternary nucleation in a system H2SO4- H2 O - NH3. All particles grow due to the condensation of gaseous H2 SO4 and organic vapours and through intra- and intermodal coagulation. The differences in results by these models are primarily due to different resolutions of particles size distribution (MULTIMONO has 4 size modes, while AEROFOR has used 27 sections) and to some extend, due to different calculation schemes for liquid water content. Largest differences are found for the smallest particles, which affect aerosol total number, but contribute negligibly to the aerosol mass.

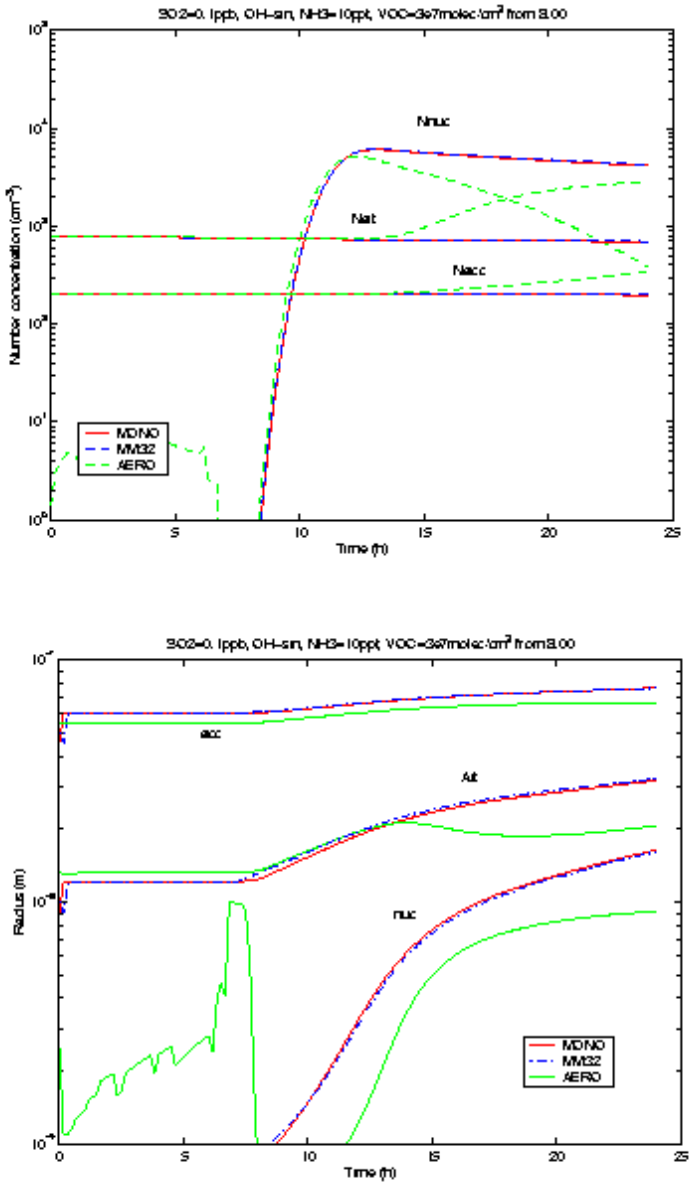

Figure 6. Evolution of aerosol number concentration and radius as calculated with MULTIMONO (MONO32 and MM32) and the sectional model AEROFOR (27 sections). (MONO32 (Helsinki University) uses FORTRAN NAG library to solve differential equations. MM32(EMEP) uses two-step time integration scheme. In this case MULTIMONO does not transfer particles to bigger bins.)

MULTIMONO is being verified against measurement data from BIOFOR 3 campaign in Hyytiala, Southern Finland, March-April 1999. Preliminary box-model calculations of aerosols number, size (Figure 7) and mass (Figure 8) for the conditions in April 14-15, 1999

(Figure 9) are presented here. The measurements (Figure 10) indicate that a nucleation event (enhanced number of very small particles) occurred at noon April, 14 followed by the particles growth. An enhanced particles growth on the second day is due to the increase in aerosol water content as relative humidity rises from 60-70% to 95-98%. In this case, MULTIMONO overestimates number of newly formed particles in nucleation mode during the nucleation burst (detection size is 3 nm in diameter). The total number concentration after 2-days simulations is quite close to the observed one. It should be pointed out that the results appear to be rather dependent on particles initial radius and number in each mode. These initial conditions have been as long as possible derived from the measurements, but due to the lack of necessary information several assumptions had to be made.

PM2.5 and PM10 concentrations (Figure 9) calculated for dry and wet (100% soluble) particles compare reasonably well with the measured PM (Figure 11) concentrations (2-days averages). Unfortunately, PM2.5 measurements are missing on very 14-15.04.

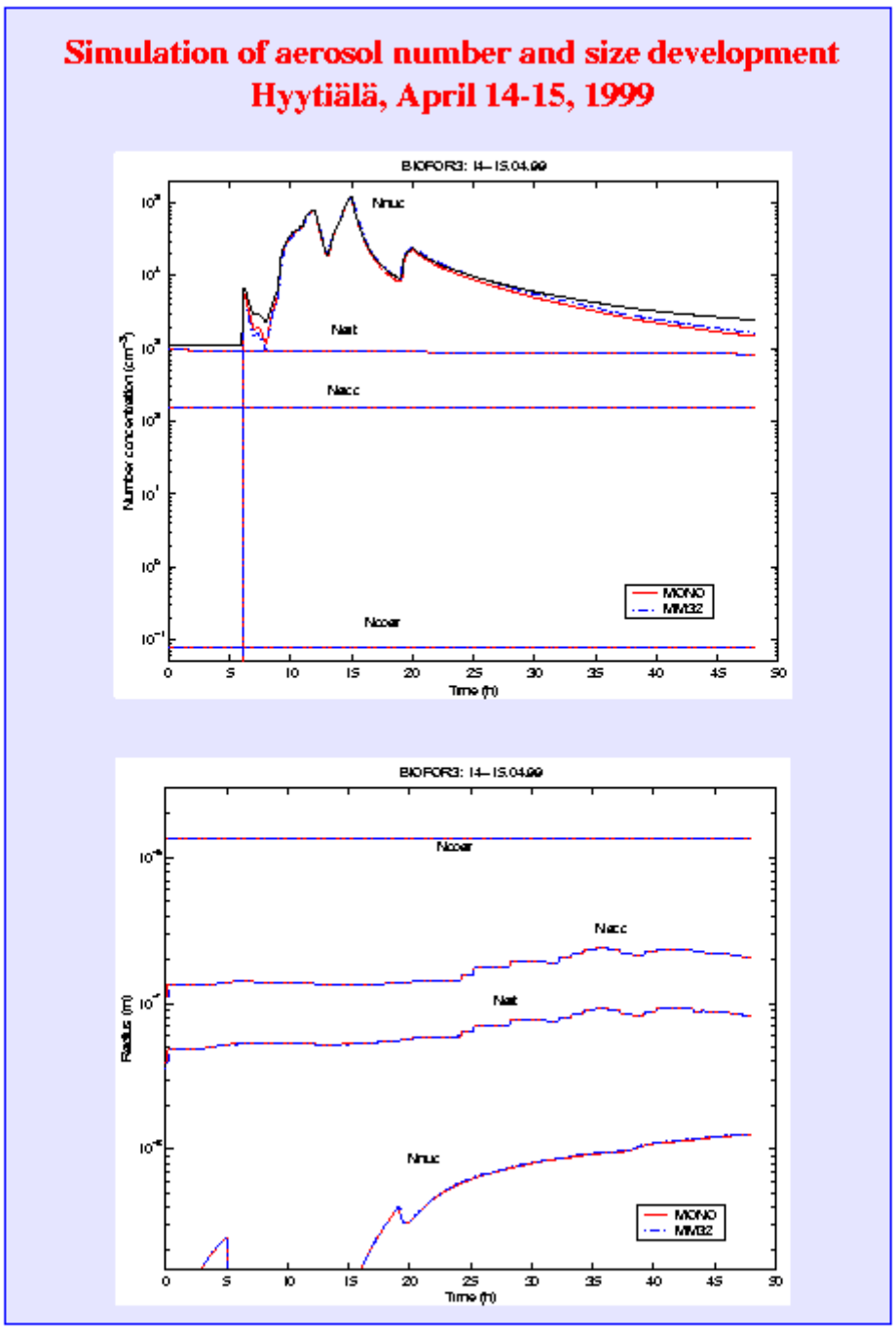

Figure 7. Preliminary box-model calculations of aerosols number [cm³] (upper) and size[m] (lower).

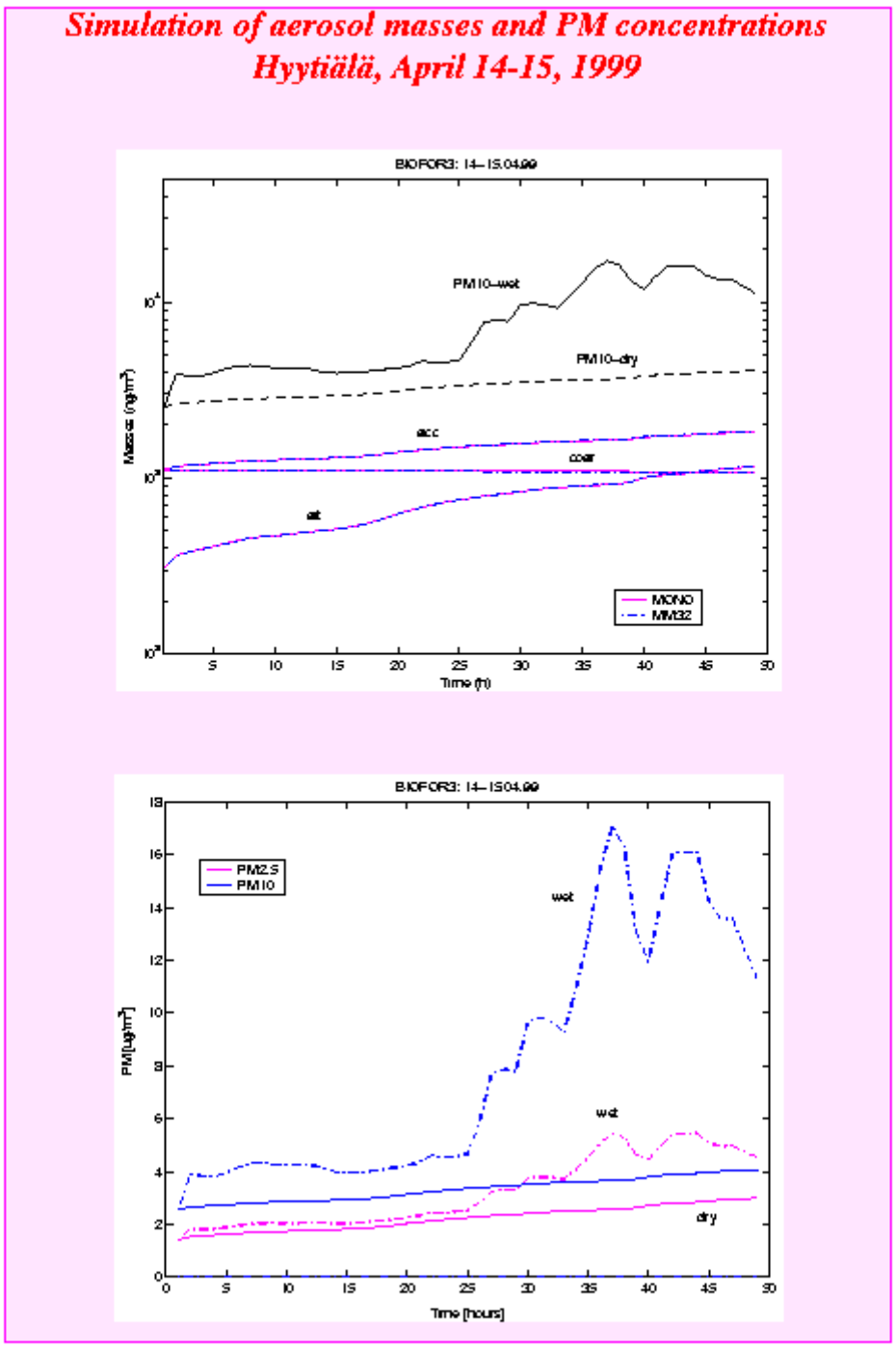

Figure 8. Preliminary box-model calculations of aerosols mass [µg/m3] (upper) and concentrations of PM25 and PM10 [µg/m3] (lower).

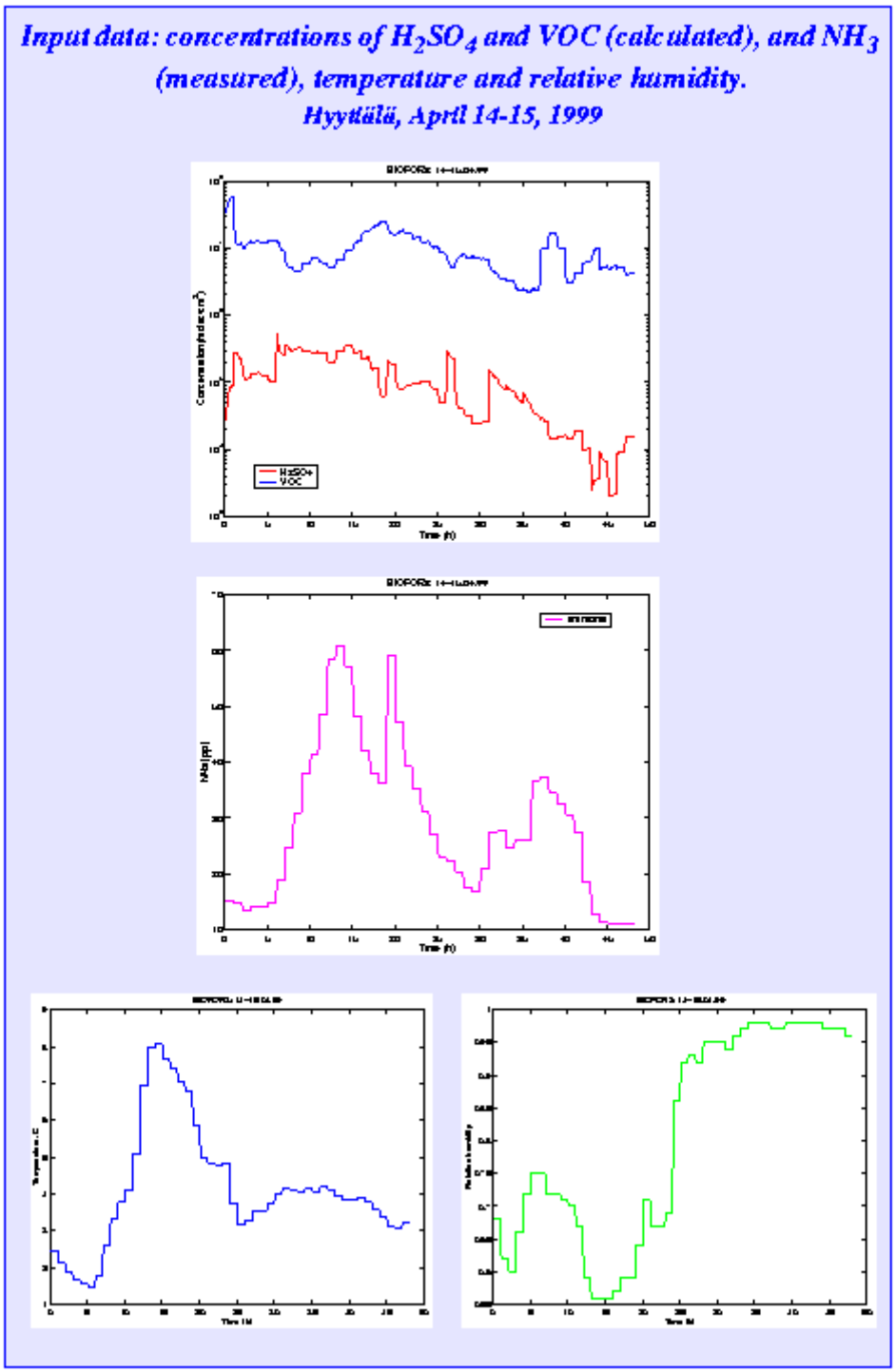

Figure 9. Conditions in Hyytiala, Southern Finland, April 14-15 1999.

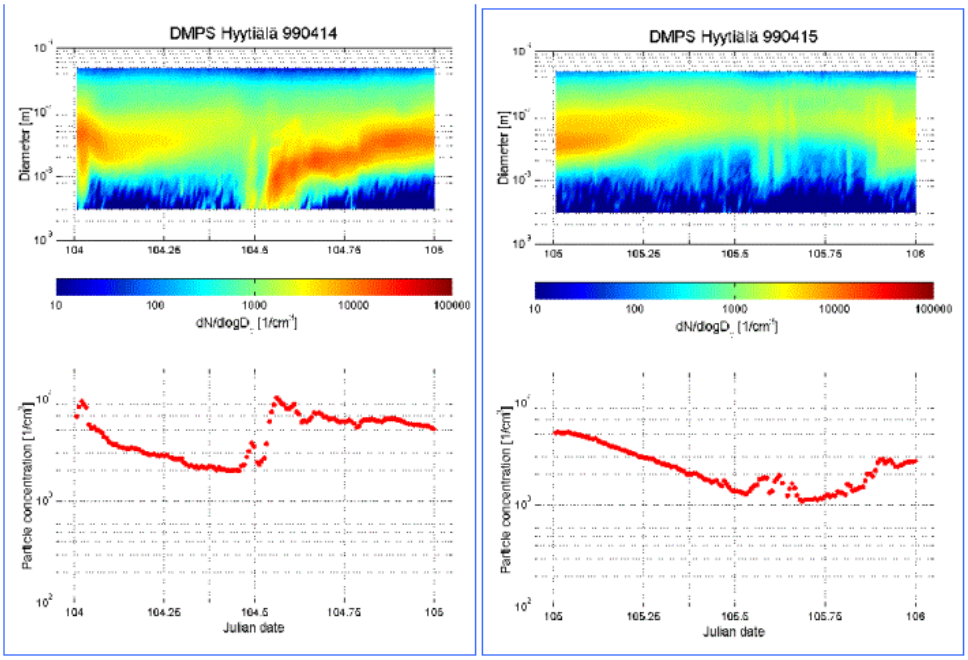

Figure 10. The development of particle size distribution and number concentration measured at Hyytiala, Southern Finland, April 14-15 1999.

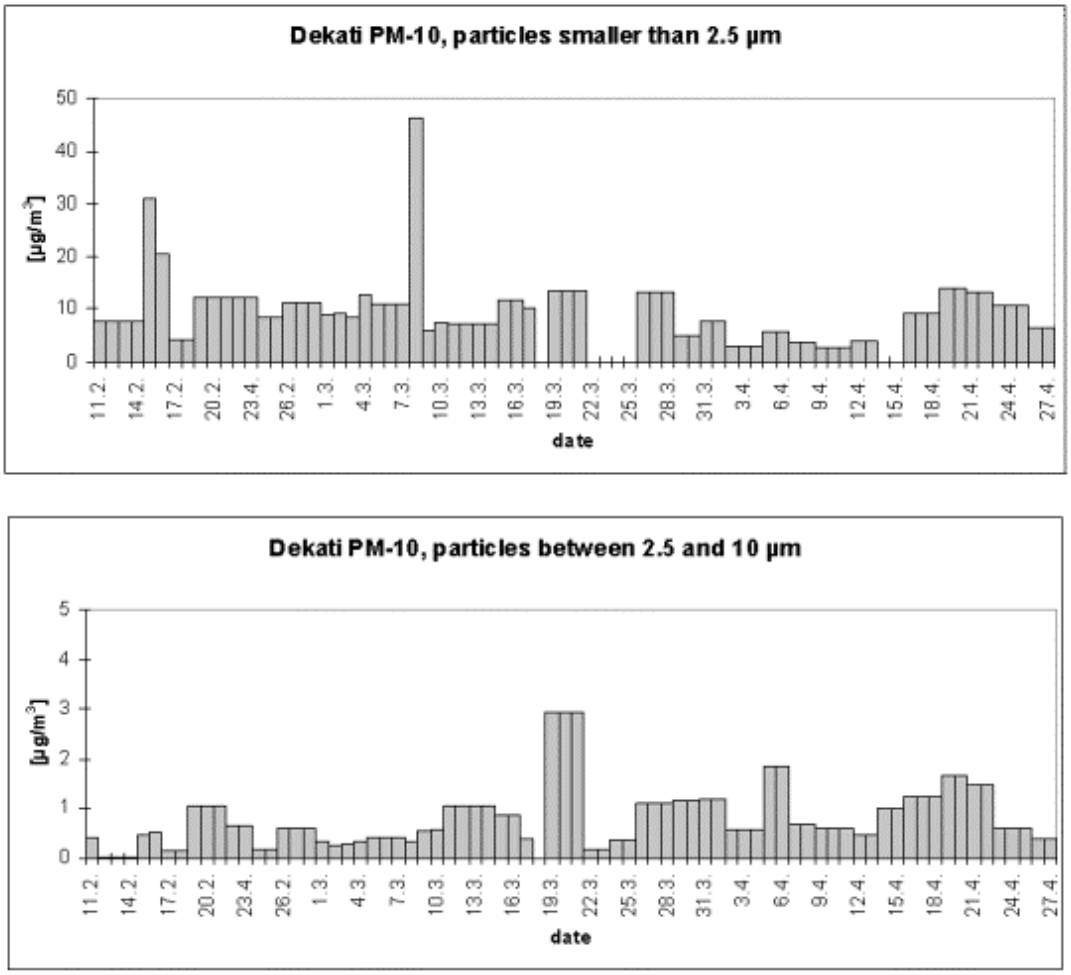

Figure 11. Measurements of the ambient aerosol mass concentration during BIOFOR3 in Hyytiala, Finland,March- April 1999.

References

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