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TNO report

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LOTOS-EUROS with M7

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Summary

Particulate matter, also known as aerosol, has an impact on climate and health. Traditionally, it was modelled in fixed size bins, but it is known that particles evolve in the atmosphere due to condensation and coagulation, which has an impact on e.g. radiative properties and the lifetime of aerosol. To include these processes in LOTOS-EUROS, the aerosol module M7 was coupled to LOTOS-EUROS and the new model version was tested.

First of all, the LOTOS-EUROS with M7 was compared to the sectional version of the model, both versions running at the default resolution (1/2×1/4˚). The models showed good correspondence for the annual mean values over Europe and time series at Cabauw. At Cabauw, results were compared with observations of mass showing a correspondence equal to that of the sectional version of the model for sodium and sulfate. Particle number concentrations were compared qualitatively to SMPS observations, as they could not be compared one to one, showing good correlations. Most particles were in the Aitken (black carbon) and accumulation (sulfate) mode, depending on the region. Furthermore, the number of nucleation events was investigated for three AERONET sites. The model underestimated the number of observed nucleation events, at Cabauw and Vavihill and strongly underestimates the number for Melpitz.

The concentrations of aerosol may have an impact on their lifetime. To study the impact of artificial dilution, i.e. the effect of horizontal model resolution on lifetime of species, the model was run at four different horizontal resolutions, from 1/8×1/16 to 1×1/2˚, for the year 2008. As expected, the annual averages were comparable but with stronger gradients and higher concentrations in source areas. Time series were again analysed at Cabauw, Vavihill and Melpitz. The correlation for the four runs per station was very high, although one could see the effect of dilution over the grid cell, in particular for Vavihill close to the coast and Cabauw in an area with strong emissions. For the highest resolution some differences are found in concentrations and dry radius of the accumulation mode at a warm and sunny day, with higher number concencentrations and slightly larger radii for the highest resolution. Overall, there is a smooth transition between the four resolutions and the effect of resolution on lifetime seems small in general. One should keep in mind that a model like LOTOS-EUROS will not be able to resolve processes at very small scale close to sources so that ageing at small scale should be taken into account already when adding the emissions to the model.

The results of the present version of LOTOS-EUROS with M7 are encouraging. Still, there are many suggestions for improvement. These include the use of a more detailed size-resolved emission database and the introduction of nitrate and ammonium aerosol. The present report should therefore be considered as a status report rather than as *the* reference document.

Contents

1 Introduction

Particulate matter, also known as aerosol, has an adverse impact on health. More and more attention is given to the smaller particles, since they can protrude deeper in the lungs and are potentially more hazardous. At the same time aerosols have an impact on climate through direct effects on the radiation budget (scattering, absorption) and via its impact on cloud formation processes. Traditionally, emission inventories of mass were produced and chemical transport models modelled the mass concentrations of species that contribute to total particulate matter (black carbon, sulfate, nitrate, ammonium, sea salt, mineral dust, semivolatile hydrocarbons and particles that are no further specified). Also observations were bases on mass concentrations. But both for health and for climate applications it is important to describe the size distribution and particle numbers.

To this end, the aerosol dynamics model M7 (Vignati et al 2004) was developed. This module takes processes like growth of aerosol due to condensation and coagulation explicitly into account. The module can be coupled to a chemistry-transport model which describes emission, transport, deposition and chemistry. It has already be implemented successfully in ECHAM (Stier et al) and TM5 (Aan de Brugh et al 2010). These are global models with a rather coarse resolution (1x1˚ lon×lat at best). The central question in the present report is whether this scale is small enough to accurately describe the aerosol processes.

To answer this question, M7 was implemented in the regional chemistry transport model LOTOS-EUROS. This model has a tradition in modelling PM (Schaap et al 2008, Schaap et al 2009, Manders et al. 2009) using a sectional approach. As a regional model, it has a finer resolution (1×1/2 up to 1/8×1/16 ˚). This report gives a full technical description of the implementation. After this technical description, the model results for the test year 2008 are discussed. They were compared with a model run using the sectional approach as a first validation. Model results were also compared with observations. These observations were both conventional mass concentration observations and particle number concentrations from the EUCAARI/EUSAAR network, with an intensive campaing in 2008. LOTOS-EUROS was used at four horizontal resolutions to investigate the impact of resolution on concentrations of mass and number.

2 M7

In the aerosol microphysics module M7, the processes of condensation of H2SO4, nucleation (H2SO4), coagulation and equilibrium with water vapour are taken into account. The module was developed and described by Vignati et al (2004).

For mass, the individual species are traced. For number, particles from different species within one mode are added. Soluble modes are in fact mixed modes, since insoluble species may become soluble due to condensation of H2SO4. When the median radius becomes too large, part of the particles is transferred to a higher mode. Note that for SO4, the number of molecules is used as a mass tracer, this is not equal to the number of particles. Densities are given in g/cm3, particle radii in cm.

Processes in M7

1. calculation of ambient mean particle mass for all modes, dry radius and density for insoluble modes, based on mass and number concentrations
2. calculation of ambient count mean radii for lognormal distribution for sulfate, mixed particles and the effect of water and sea salt
3. calculate effect of condensation, nucleation and coagulation, change particle-number relationship
4. recalculation of particle properties under ambient conditions (point 1 and 2)

Table 2.1 Definition of modes as used by M7

Mode	dry radius (μm)	Geometric stdev σ	components
1 Nucleation soluble	0.0005-0.005	1.59	SO4
2 Aitken soluble	0.005-0.05	1.59	SO4, BC,OC
3 Accumulation soluble	0.05-0.5	1.59	SO4,BC,OC, SS, DU
4 Coarse soluble	>0.5	2.0	SO4, BC,OC, SS, DU
5 Aitken insoluble	0.005-0.05	1.59	BC, OC
6 Accumulation insoluble	0.05-0.5	1.59	DU
7 Coarse insoluble	>0.5	2.0	DU

3 Implementation of M7 in LOTOS-EUROS

LOTOS-EUROS is a Eulerian chemistry transport model. It is used on the European domain, from 10˚W-40˚E, 35-70˚N on a 1/2×1/4˚ longitude-latitude grid with 4 dynamical vertical layers, including a 25 m surface layer, a mixing layer and two reservoir layers.

Modelled species are ozone, nitrogen oxides, ammonia, primary PM2.5 and black carbon, primary PM10 (excluding PM2.5 and black carbon), sulfate, nitrate, ammonium and sea salt and species relevant as precursors or reservoir (per-acetylnitrate, volatile organic carbon). For (photo)chemical gas reactions the CBM IV scheme is used, for secondary inorganic aerosol (heterogeneous chemistry) Isorropia or EQSAM can be used.

LOTOS-EUROS with M7 takes into account the primary emissions of aerosol and their subsequent transport, deposition (wet and dry) and the aerosol processes of nucleation, condensation and coagulation, effect of relative humidity. Particles may go from one mode to another mode (insoluble to soluble, larger mode). The gas-phase chemistry of LOTOS-EUROS may be used to calculate the production of H2SO4. In the present set-up chemistry resulting in the formation of secondary organic or inorganic aerosol is not coupled to the M7 species and processes.

The implementation was based on LEv1.5 with patch 026 and project bestguess (horizontal diffusion switched off). The new code was implemented in project LEM7-new and appendix A describes how to run the model with M7 and the output variable names and units.

To translate the LOTOS-EUROS arrays to M7 compatible arrays, the module callm7.F90 was written, containing the subroutine m7interface. Mass and number concentrations are treated like the other concentrations in the transport and deposition routines of LOTOS-EUROS. Other properties of the distribution like density and radius are not transported in LOTOS-EUROS.

In LOTOS-EUROS, M7 is treated as an extension to the chemistry. The chemistry may be used to calculate the H2SO4 production. Thus, the call to M7 is directly after the call to the chemistry routine in the operator splitting.
However there are processes in Lotos-Euros which affect the size distribution and some are in turn dependent on the size distribution. Therefore, an extra call to the M7 routines which recalculate the radius based on the instantaneous mass and number concentrations should be made before the second call to the dry deposition routine in the operator splitting. Processes like advection and emission do have an effect on mass and number concentrations and in this way the radius of the distribution, but do they do not explicitly depend on the radius of the distribution. Therefore, it is not necessary to recalculate the radius after each process. But an update of radius and density is necessary before the deposition is calculated. To prevent the unphysical case of a grid cell would containing mass and number but particles have no radius or density an update of density and radius can be made trough a call to the subroutine updateaerosol.F90 (subroutine updatemodes). This is an interface to the specific M7 routines which recalculate the distribution.

An artefact that may arise due to specific choices made in M7 is that no mass and number, but finite density and radius for SO4_nuc appear after a call to M7 when no SO4 is present. This is due to a specific choice in m7_averageproperties which is made to be able to calculate the intramodal coagulation coefficient.

3.1 Emissions

As a basis for the emissions the TNO gems emission database was used, which includes ozone precursors and primary anthropogenic PM.

3.1.1 H2SO4

The concentration H2SO4g can be calculated in the chemistry scheme of LOTOS-EUROS. Only the reaction between SO2 and OH is taken into account presently:

c(H2SO4)=c(H2SO4)+CF*RK*c(SO2)*c(OH)

where the conversionfactor CF includes the timestep and the conversion to molecules/cm3 and the reaction constant RK is calculated in LOTOS-EUROS (rk(71)).

3.1.2 Cloud processing

Cloud processing of SO2 is an important source of sulfate. It contributes more than the formation of H2SO4. The process was parameterized by the reaction

c(SO4_ait)=c(SO4_ait)+CF*RK2*c(SO2).

Analogous to H2SO4, a conversion factor CF is used. The reaction constant RK2 depends on cloud cover and relative humidity. The resulting sulfate is assumed to go into the Aitken mode.

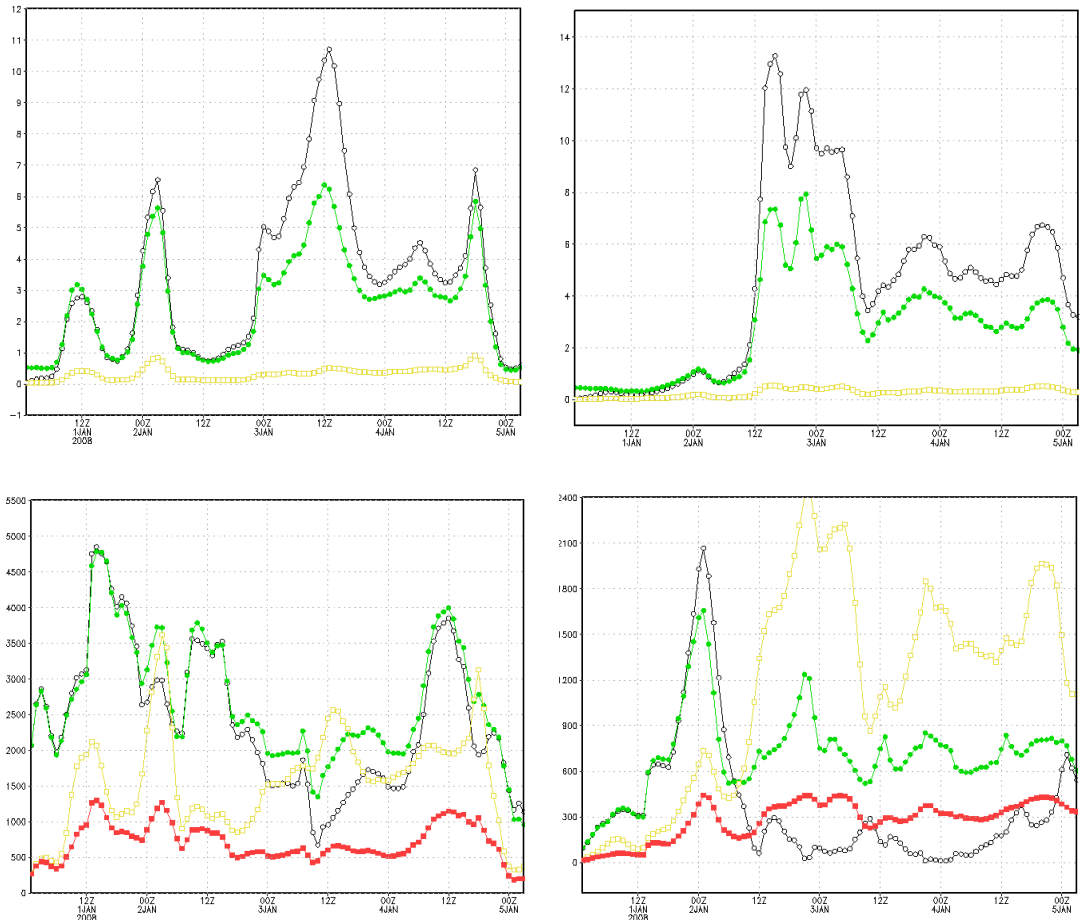

Figure 1 Upper panels: total SO4a in ug/m3, Cabauw (left) and Melpitz(right). Black: LE_m7, green: LE_sect, yellow: LE_m7 without cloud processing . Lower panels: particle numbers, #/cm3, Cabauw (left) and Melpitz(right). With cloud processing: Black: aitken, yellow accumulation, without cloud processing: green: aitken, red accumulation

3.1.3 Aerosol emissions

Primary emissions of SO4 and BC can be taken from one of the emission databases available for Lotos-Euros. These include annual mass emissions and time profiles. These mass emissions must be divided over the appropriate M7 mass modes and the corresponding number emissions must be calculated. In emis_mod.F90 (subroutine mkemis) these emissions are read (or constructed, for sea salt and dust). In subroutine emism7 these are translated into the emissions with suit the conventions of Lotos-Euros with M7. This routine is called from subroutine addemis.

Choices for the attribution to the modes, their density and their mean radii are based on the choices made for the emissions in TM5, see table 3.1, but without the discrimination to source category (fossil fuel, biomass burning, anthropogenic or industrial emission, volcanoes). This can be improved upon, but this also requires in part a different emission database. In mo_aero_m7.F90 also some densities are set, they can be used directly (after a unit conversion) but this was not implemented here. In principle it is thus possible to introduce an inconsistency.

- SOx emission: 2% of the SOx emissions are assumed to be emitted directly as aerosol. The mass of this aerosol is divided equally over the Aitken and the accumulation mode.
- BC emissions: these are based on predefined fractions of the PPM2.5 emissions in LOTOS-EUROS. The mass and number are fully attributed to the Aitken insoluble mode
- OC emissions: not in LOTOS-EUROS emission database. As a crude approximation, the BC emissions are taken as a proxy: OC=3*BC. This is an average, in fact the ratio changes with season and location: on traffic-oriented sites OC=1.1*BC is more appropriate whereas in remote rural sites OC=8*BC may be found (Gelencser et al 2004, Schaap and Denier van der Gon 2004). The OC is further subdivided, with 65% in the soluble and 35% in the insoluble Aitken mode according to the choice made in TM5.
- SS emissions. The basic sea salt emission routine with Monahan source function and a division in the sectional size classes 0.14-2.5 and 2.5-10 um was used. The emission and sea salt routine were adapted to avoid conflicts with array dimensions and indices. Since these size classes are for a wet diameter at 80% relative humidity, they correspond to dry radii of 0.035-0.625 and 0.0625-2.5 um, which makes the translation into the LOTOS-EUROS accumulation and coarse modes reasonable as a first guess. Note that only sodium is used as a tracer, for total sea salt the chloride mass (and other elements) should be added to the mass.
- Dust emissions are not yet fully available in LOTOS-EUROS and were not taken into account in the present project. They can in principle readily be included in the emission structure.

To calculate the number emission from the mass emission:

$$r_{avmass} = r_{medianmass} \exp(1.5 \ln^2 \sigma)$$

$$number = mass * \frac{3}{4\pi r_{avmass}^3 \rho}$$

Note that M7 does not check for consistency between mass and number emissions, since mass and number together with density determine the radius, and the number is a sum of the numbers of different species.

Table 3.1 Density and median radius used for the emissions of primary aerosol

species	density (kg/m3)	median radius (um)
SO4_ait	1841	0.03
SO4_acc	1841	0.075
BC_aiti	2000	0.034
OC_aiti	2000	0.025
OC_aits	2000	0.025
SS_accs	2165	0.079
SS_coas	2165	0.63
DU_accs	2650	0.079
DU_coas	2650	0.63
DU_acci	2650	0.079
DU_coai	2650	0.63

3.2 Dry deposition

For dry deposition a hybrid approach is used, based on the existing deposition and sedimentation routines in LOTOS-EUROS to calculate the change in concentration and fluxes. The calculation of sedimentation and deposition velocities is partly based on the approach taken in TM5, partly on the routine rb_zhang. The latter was developed for LOTOS-EUROS and is explicitly dependent on the radius. Sedimentation and dry deposition velocities are calculated in the module drydepom7.F90. Velocities are calculated for both mass and number separately, resulting in a change in the radius of the distribution.

3.2.1 Sedimentation

The sedimenation velocity is calculated in the subroutine vsed.
For sedimentation of number the number median radius (first mode) $\overline{D}_{pgR} = \overline{D}_{pg} \exp(\ln^2 \sigma)$ is determined.
For sedimentation of mass the mass median diameter (third mode) $\overline{D}_{pgV} = \overline{D}_{pg} \exp(3 \ln^2 \sigma)$ is used.
The sedimentation velocity (Stokes velocity) is then calculated using these mean radii. For sedimentation of mass an additional factor $\exp(2 \ln^2 \sigma)$ (Slinn factor?) was applied to the sedimentation velocity. In depos.F90 these sedimentation velocities are applied to mass and number concentrations like for the other LOTOS-EUROS aerosol species.

3.2.2 Dry deposition

For dry deposition the velocities are not simply based on median radius but on a convolution of the mode with the function describing deposition velocity as a function of radius. The approach taken in TM5 (Maarten Krol) is followed for the

deposition of mass and number, including convolution with a look-up table, and deposition velocities based on Zhang (2001).

First a look-up table is constructed for deposition velocities (subroutine depotable), including 23 particle radius bins varying from 0.001 to 100 um, using a reference density of 1800 kg/m3. The actual deposition velocities for mass and number are calculated in the routine deposvelpart by applying a convolution of the aerosol distribution with the table. Before applying this convolution the look-up table is corrected for the actual density of the aerosol mode, which may introduce a shift in the table.
The deposition velocy per mode is calculated by convoluting the distribution

$$n_N(D_p) = \frac{N_t}{\sqrt{2\pi}\, D_p \ln \sigma_g} \exp\left(- \frac{(\ln D_p - \ln \overline{D}_{pg})^2}{2 \ln^2 \sigma_g} \right) \text{(number)}$$

$$n_V(D_p) = \frac{\pi}{6} D_p^3 n_N(D_p) \text{ (mass)}$$

with the table with deposition velocities $v_d(D_i)$ so that the deposition velocities are

$$v_{dN} = \frac{\sum n_N(D_i) v_d(D_i) (d \ln D)_i}{\sum n_N(D_i) (d \ln D)_i} \text{ (number)}$$

$$v_{dN} = \frac{\sum n_V(D_i) v_d(D_i) (d \ln D)_i}{\sum n_V(D_i) (d \ln D)_i} \text{ (mass)}$$

In the routine mix2ground these velocities are also used to translate the concentrations from model level 1 to observation level (ground level). Note that in the routine le_output.F90 the update of deposition velocities is disabled for consistency and efficiency reasons.

In M7, aerosol may grow into the next size class, but it does not take into account a shrinking of the particles. The current deposition scheme may result in radii that are lower than the official lower limit of the size class for coarse particles at very low concentrations. Since mass and number are both used as a tracer, as a consequence the radius is allowed to vary. In the deposition routine, the deposition velocity was set to 0 for particle number concentrations smaller than 0.1 particle/cm3 to circumvent too unrealistic behaviour. The consequence is that low background mass concentrations of the order of 0.1 ug/m3 can be present for coarse particles like sea salt and dust. However, as this version of LOTOS-EUROS is aimed at modelling particle number and particle interactions, this is not a serious drawback.

3.3 Wet deposition

A simple approach is used, based on the choices in EMEP UNI-AERO. This is consistent with the LOTOS-EUROS approach which also uses EMEP wet deposition parameters. For mass and number the same below-scavenging efficiencies are used, no discrimination is made between soluble and insoluble and the mode median radius is not taken into account. This is a crude approximation,

but since scavenging efficiencies are rather poorly constrained it is difficult to improve on this. TM5 uses different scavenging efficiencies but also treats mass and number equally. Parameter values for the below-loud scavenging efficiencies E are given in Table 3. Only below-cloud scavenging is taken into account. When in-cloud scavenging is implemented in Lotos-Euros, it is easily extended for aerosol modes analogous to the below-cloud scavenging using the wash-out ratios W in Table 3.

Table 3.2 Washout ratio and Scavenging efficiency

	nucleation	Aitken	accumulation	coarse
Washout ratio W (*10^6)	0.0	0.2	0.7	0.7
Scavenging efficiency E	0.4	0.3	0.1	0.4

4 Model results

4.1 Approach

First of all, several box tests were performed to test the implementation of M7 and the impact of deposition. Subsequently, plume tests were carried out to get an impression of the combined effect of aerosol dynamics, deposition and transport. These experiments will not be discussed here.

Simulations were performed over the year 2008, which is the year for which an intensive European field observation campaign was done. A model run of the sectional version of LOTOS-EUROS was done, as well as a run with the version with M7, both at the model's standard resolution. In addition, a run at lower resolution and two runs at higher resolution (zoom runs) were performed, with the zoom runs using boundary conditions of tracer mass and number concentrations from the run at standard resolution:

- Domain 10W-40 E, 35-70N, 1.0x0.5, coarse
- Domain 0-15 E, 48-60N, 0.25x0.125, zoom factor 2
- Domain 3-9 E, 49-55 N, 0.125x0.0625 zoom factor 4

The zoom domains were chosen such as to cover the densely populated areas in Northwestern Europe and part of the North Sea, but avoid mountain regions for which LOTOS-EUROS may perform less well. The Netherlands has been chosen as the central region for the maximum zoom resolution, since it is an area with high anthropogenic PM concentrations (Randstad, Ruhr area) but is also influenced by sea salt aerosol and shippinig emissions and has a rather flat topography.

The zoom factor 2 regions contains the EUCAARI stations Cabauw, Melpitz and Vavihill (www.eusaar.net, Manninen et al 2010). Cabauw is a rural station situated rather close to areas with high emissions, which can also be influenced by clean maritime air. Melpitz is a rural station far away from emission hotspots. Vavihill is a station in a rural area rather close to the sea, with clean air coming from the northern directions, but also influenced by nearby cities in the southwest.

First, the annual averages were compared with averages for the sectional model version, then the distribution of the species over the modes will be discussed. After that, annual averaged results from the zoom runs are presented and compared with results for the standard resolution. Then time series will be shown and discussed at Cabauw, Melpitz and Vavihill and compared with observations when possible. Finally we try to sketch a unified picture by considering transport and vertical structure.

4.2 Annual averages

4.2.1 Comparison with sectional LOTOS-EUROS results

As a next step, the full year 2008 is simulated with LOTOS-EUROS including M7, with emissions as described above, and zero boundary conditions. This simulation is compared with a traditional LOTOS-EUROS simulation, with a sectional approach (v1.6.2). Only total mass can be compared, and one should keep in mind that the

deposition in the traditional model is different (depac) which may introduce rather large differences, in particular for the coarse particles.

First of all, the annual averages of sea salt, black carbon and sulfate mass concentrations were compared at ground level (Fig. 2). For sulfate and black carbon, annual averages are comparable with the sectional approach. For sea salt the concentrations in the M7 version are higher, but this is an effect of the Zhang deposition scheme as compared to the Depac deposition (Schaap et al 2009).

4.2.2 Distribution of species over the modes

In figures 3-6 the distribution of the species over the different modes and total particle number concentrations at ground level are shown. It is not easy to interpret the map of nucleation mode. High concentrations are found for mountain regions, for which the results of LOTOS-EUROS are less reliable. This mode will discussed in more detail when the effect of resolution is treated and series at station level are discussed. Concentrations seem not related to the H2SO4 concentrations, for which concentrations are suspicioulsy high near the boundaries of the LOTOS-EUROS domain. This might be the effect fo using zero boundary conditions, so that near these boundaries there is not enough aerosol to condense on. For the aitken mode, emission hotspots of carbon and SO2 (in part directly as sulfate) can be identified like the ship tracks, the Randstad area (Netherlands), the Ruhr area (Germany) the Po valley (Italy), major cities (Paris, London, Madrid) and industrial aeras in the East stick out. Concentrations are highest in the accumulation mode, consisting partly of direclty emitted species and partly of aged particles from the Aitken mode. Due to this ageing the pattern of emissions gets blurred by the transport. In the accumulation mode, the Balkan (in particular Bulgaria and Romania) and eastern Turky stick out. The concentration in the coarse mode is low. This mode consists purely of aged aerosol, which results in low gradients with slightly elevated concentrations in southern France and northern Italy and again in Turkey.

The black carbon particles are mainly in the insoluble aitken mode, the mode in which they are emitted, and the soluble accumulation mode, the mode in which they arrive after ageing. Some particles are in the aitken soluble mode but the concentration is very low, and they do not (or hardly) grow into the coarse mode. The emission hotspots are clearly visible for the aitken mode, and the accumulation mode is a blurred version of this pattern due to transport.

For sea salt most of the mass is in the coarse mode, although for inland conditions the difference decreases.

Particle number distributions clearly show the emission hotspots of sulfate and black (and organic) carbon in the Aitken mode and a blurred version in the accumulation mode (with lower particle numbers as the particles are larger but heavier). The coarse mode reflects the pattern of sea salt concentrations. Particle numbers of the Aitken and accumulation mode are in line with observations (UFIPOLNET). In the time profiles, mass concentrations with M7 are slightly larger than with the sectional approach for the periods with relatively high concentrations. Total particle numbers are in general agreement with particle number

concentrations reported in the literature, except for the nucleation mode, for which the variability in time is quite high.

These results indicate that the implementation of M7 gives results which are in line with the sectional version of the model and in line with observations of particle numbers.
This implies that we can do zoom experiments to investigate the impact of horizontal resolution.

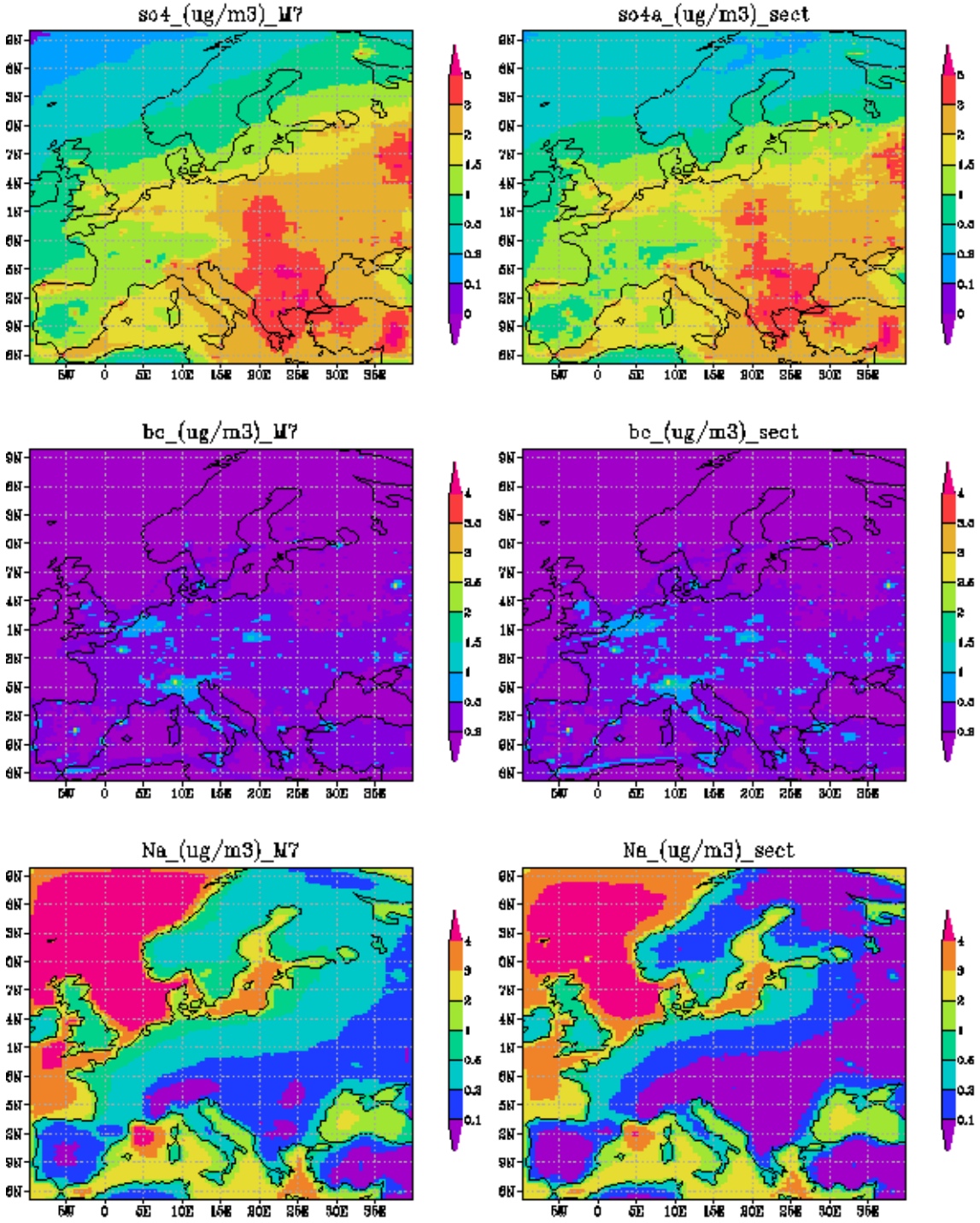

Figure 2 Annual averages of mass concentrations of sulfate, black carbon and sodium, simulated for 2008. Left: LOTOS-EUROS with M7, right: sectional LOTOS-EUROS

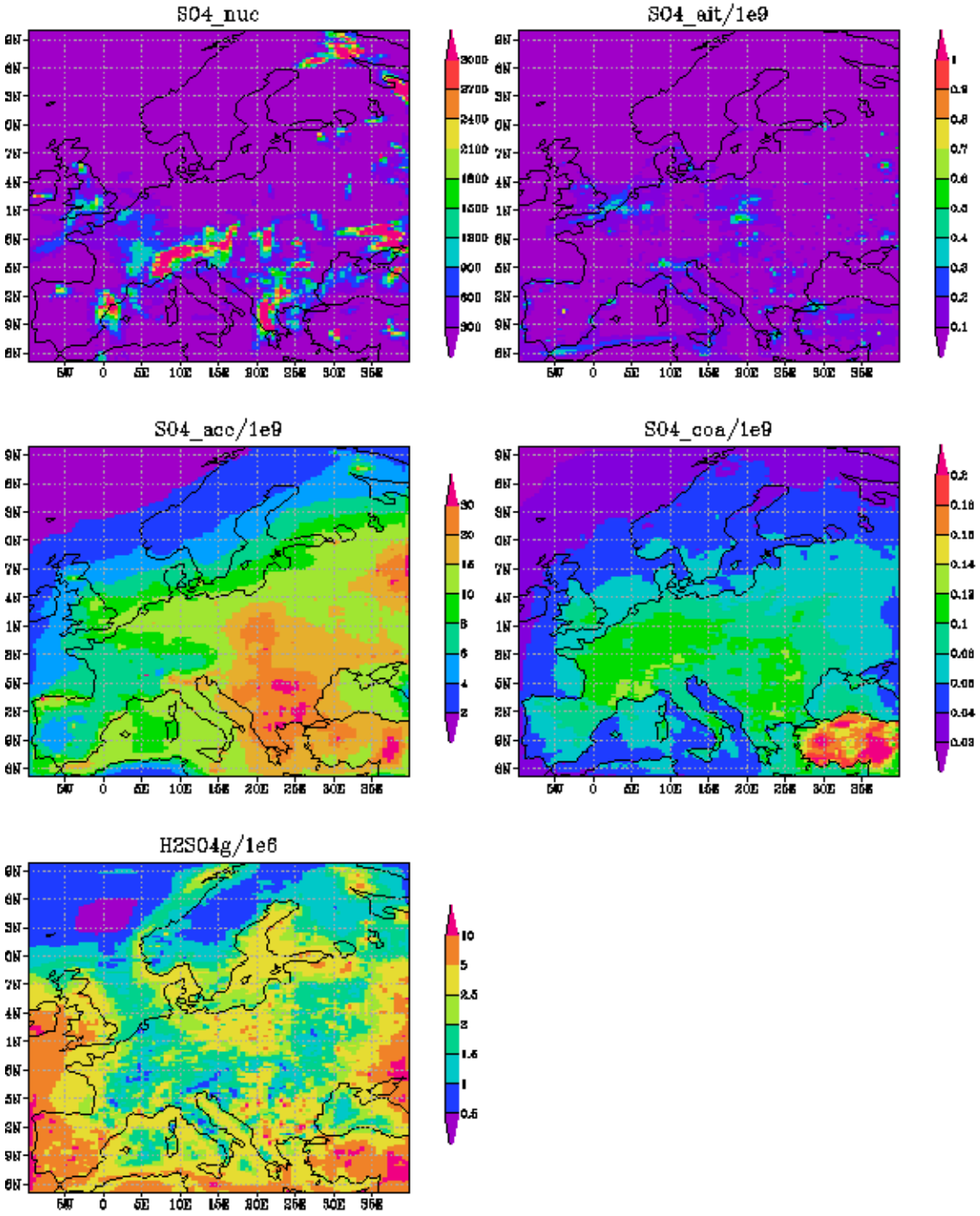

Figure 3 Annual averages of sulfate concentrations, 2008, in 10^9 molecules/cm^3 and H2SO4 (g) in 10^6 molecules/cm^3

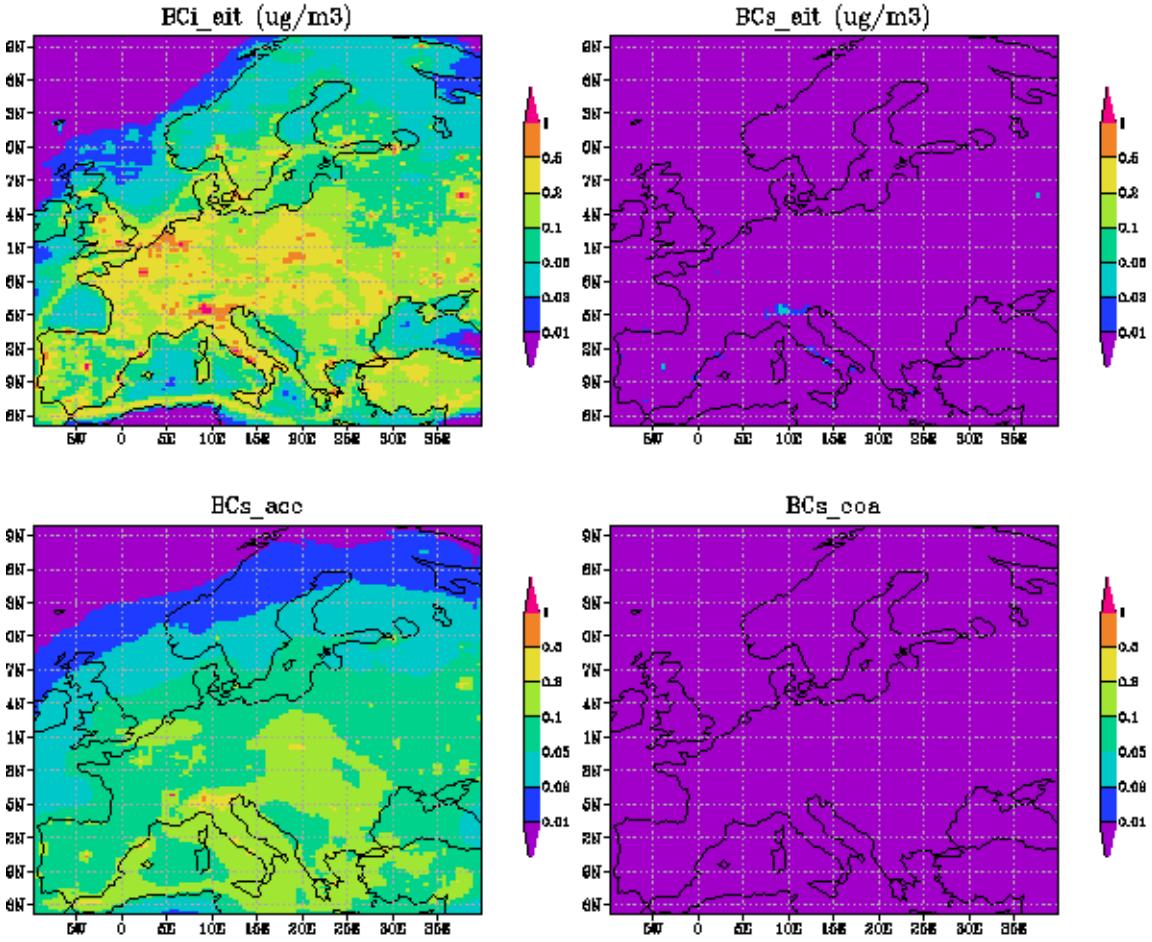

Figure 4 Annual averaged BC concentrations, 2008, µg/m3

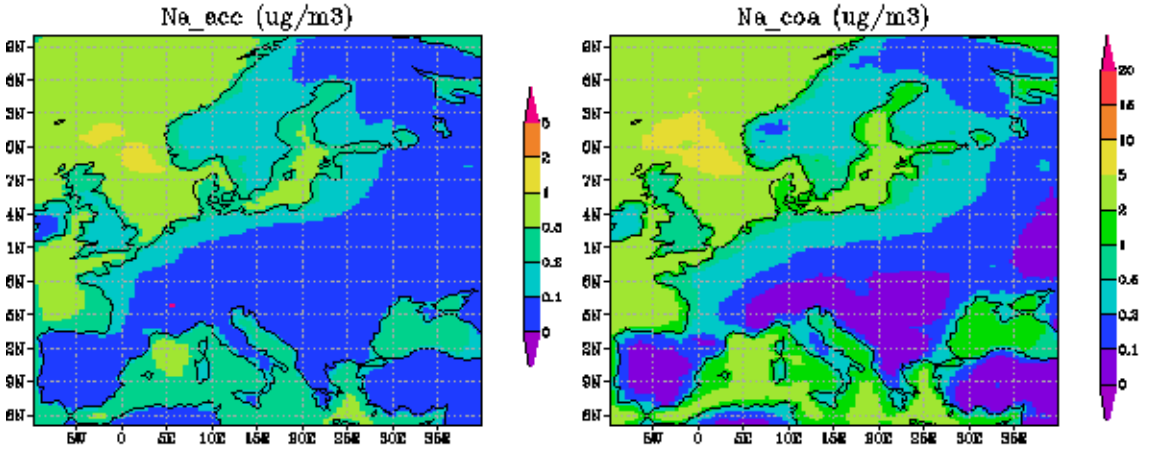

Figure 5 Annual average mean sodium concentrations 2008, µg/m3

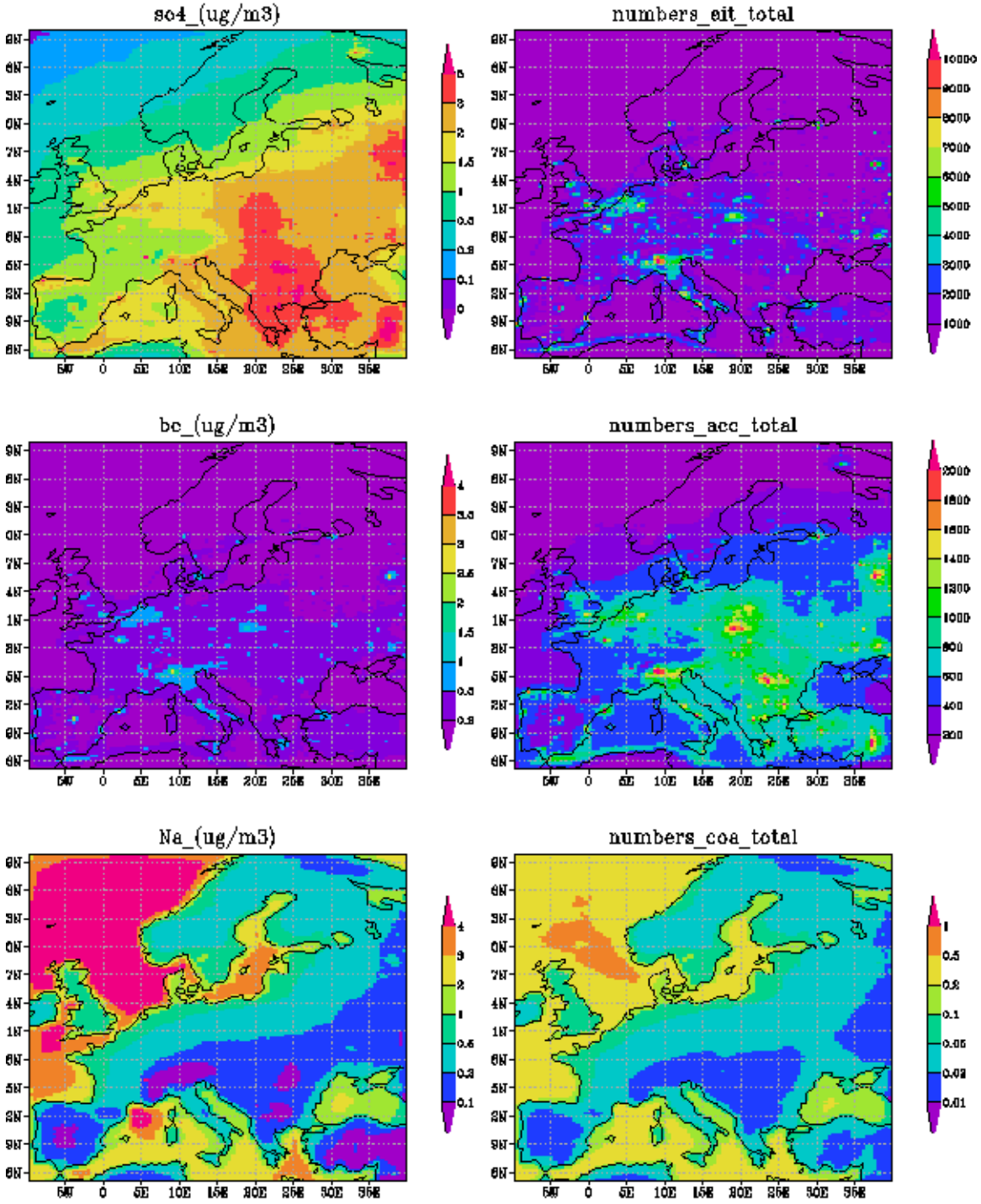

Figure 6 Mass and particle number concentrations, annual averages 2008

4.2.3 Runs with different resolution

The annual average mass and number concentrations for the three resolutions (Figs 7-12) do not differ significantly from each other. Concentrations are somewhat higher at higher resolutions for specific source regions and the patterns show more detail, as expected. Differences are largest for the nucleation mode, note fore example the differences around the Eifel region (Fig. 11). This indicates that the lifetime of the species is on average not different for the different resolutions.

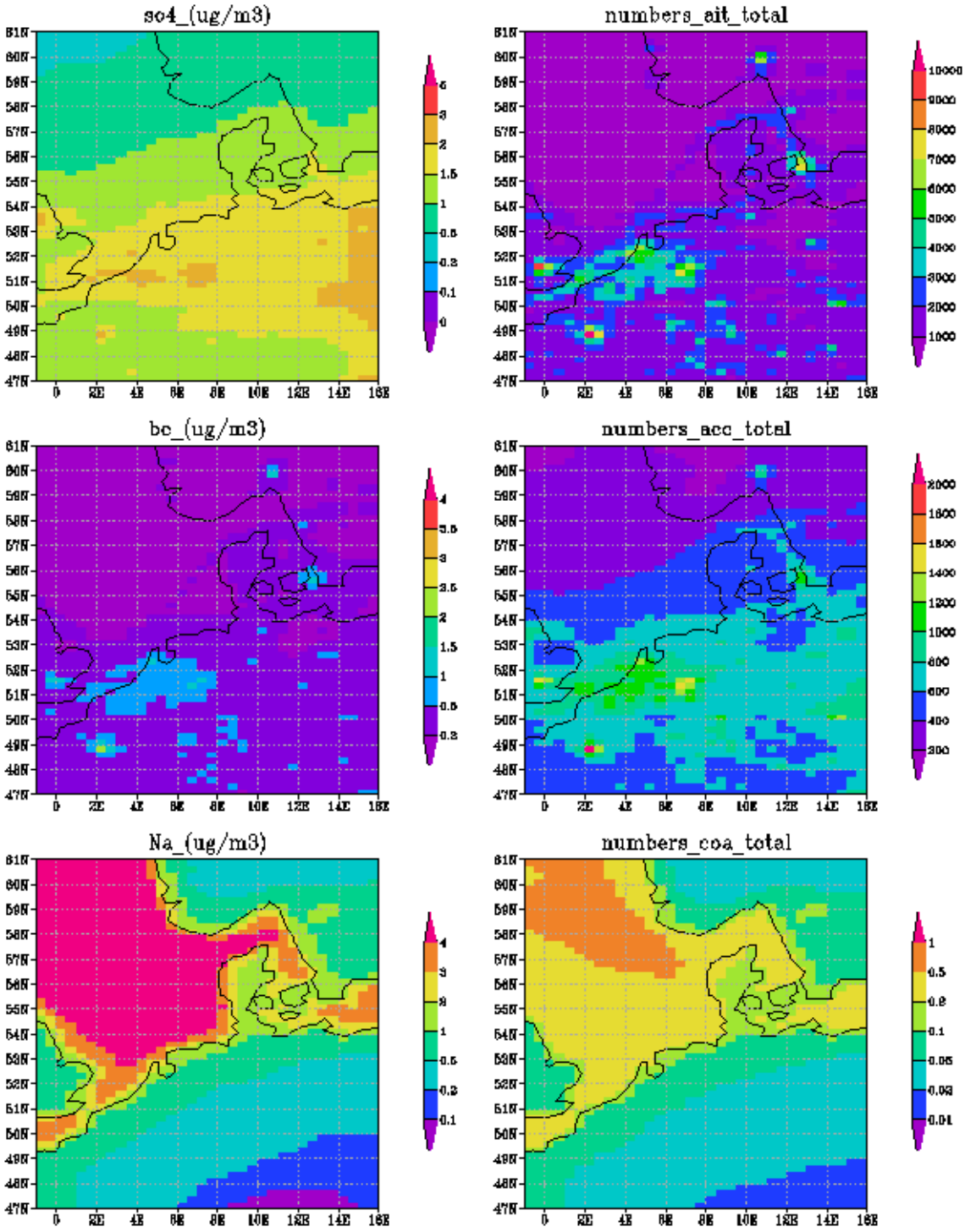

Figure 7 Concentrations in zoom domain, standard resolution (0.5x0.25)

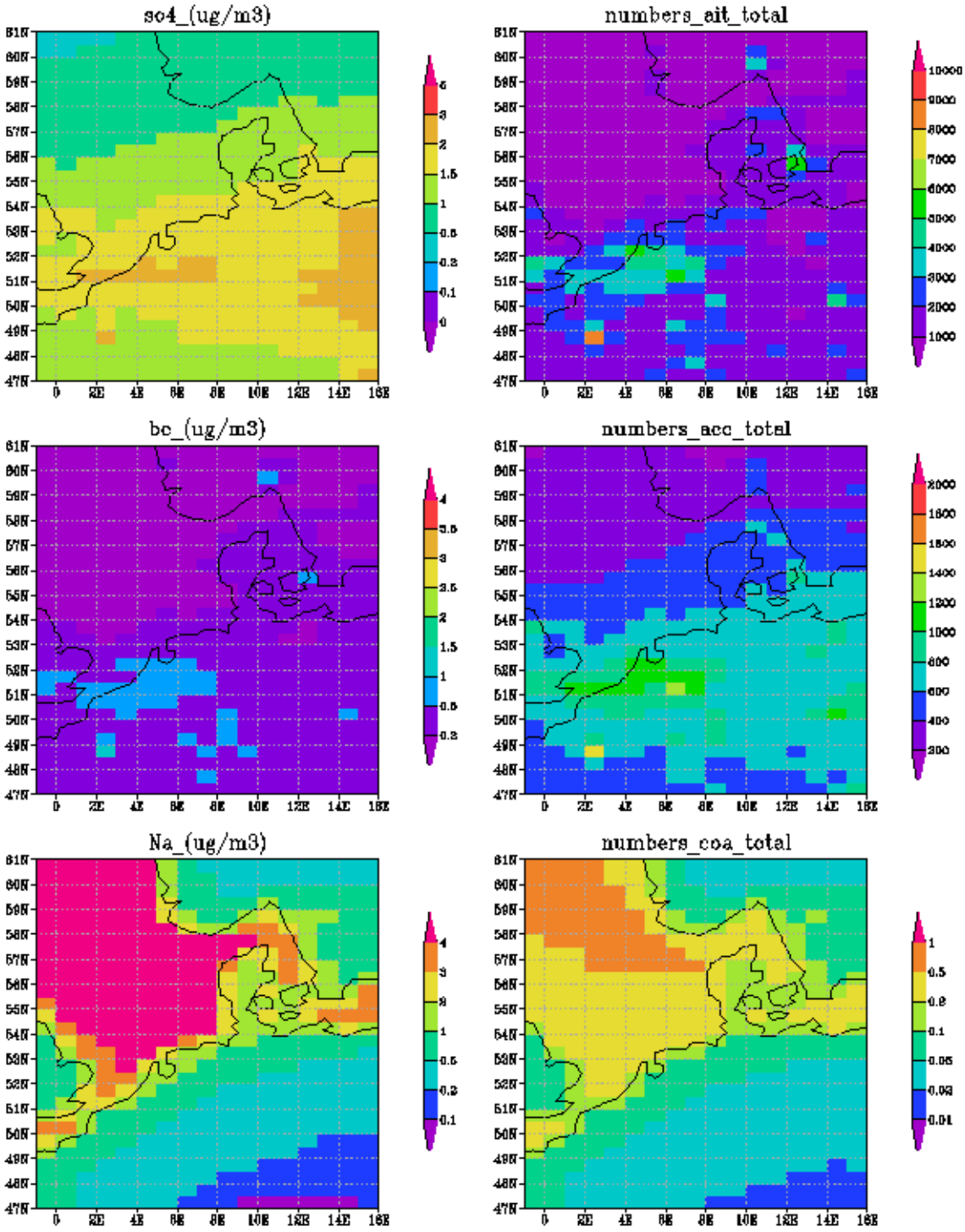

Figure 8 Concentrations in zoom domain, coarse resolution (1x0.5)

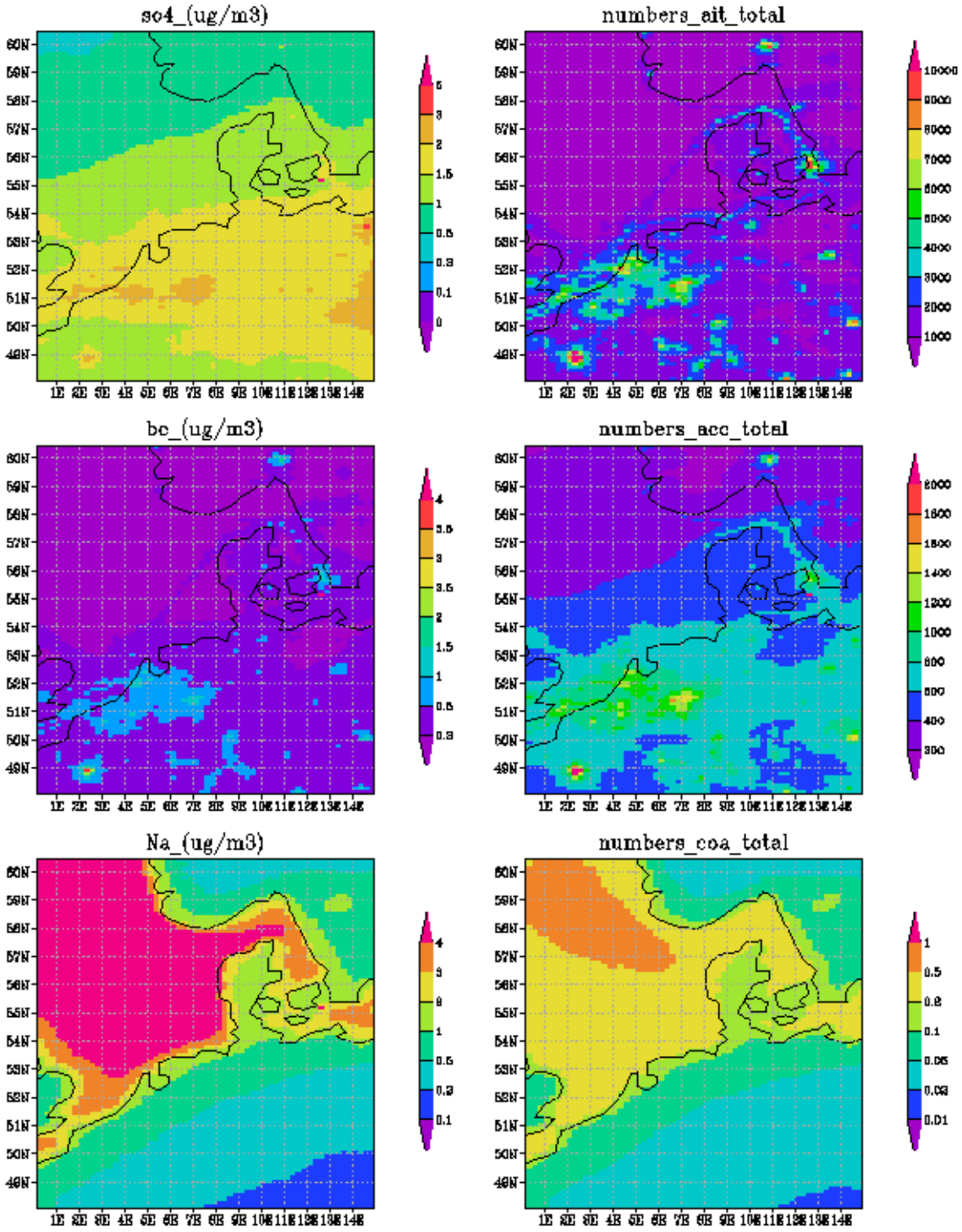

Figure 9 Concentrations in zoom region, higher resolution (0.25x0.125)

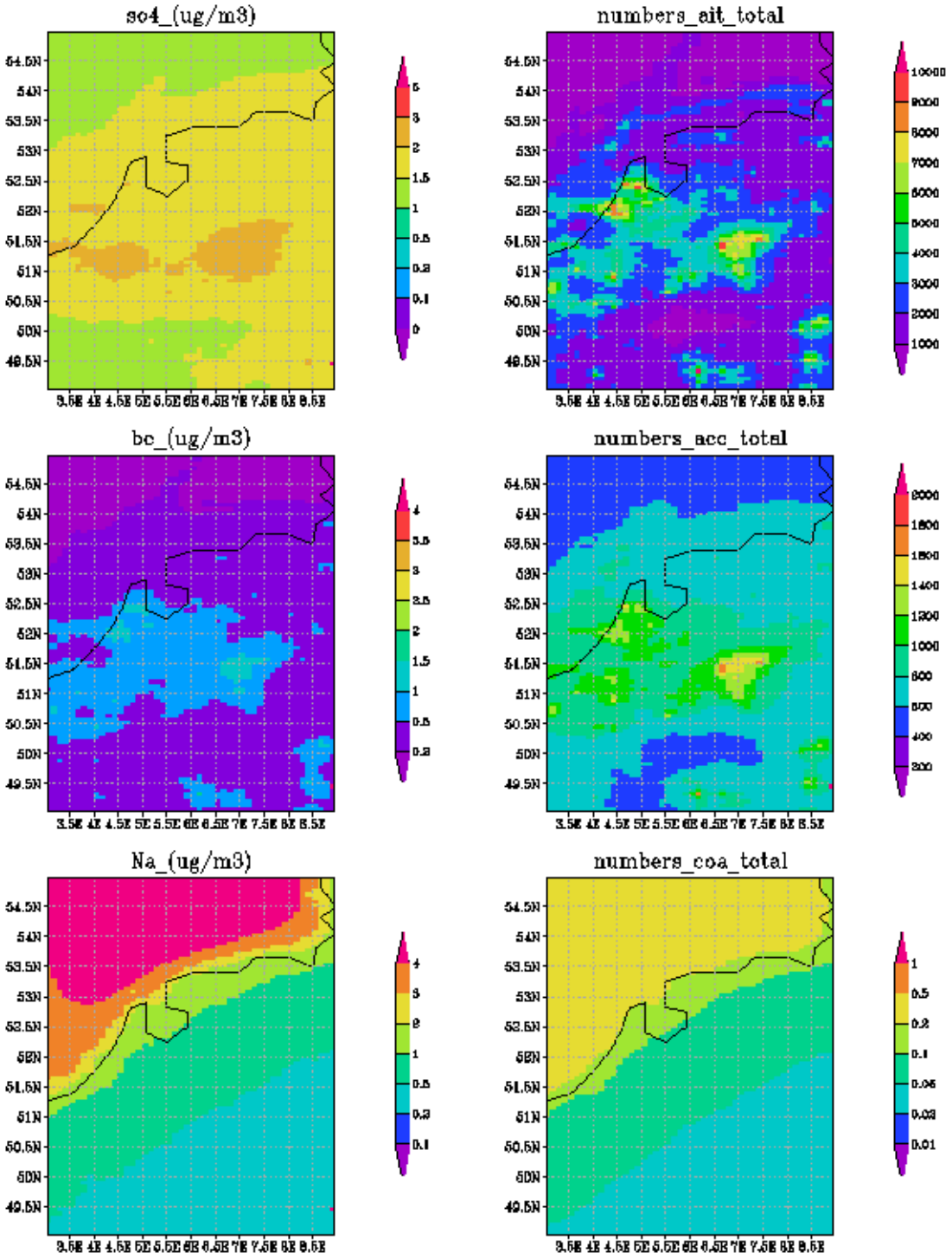

Figure 10 Concentrations in zoom region, highest resolutions (0.125x0.0625)

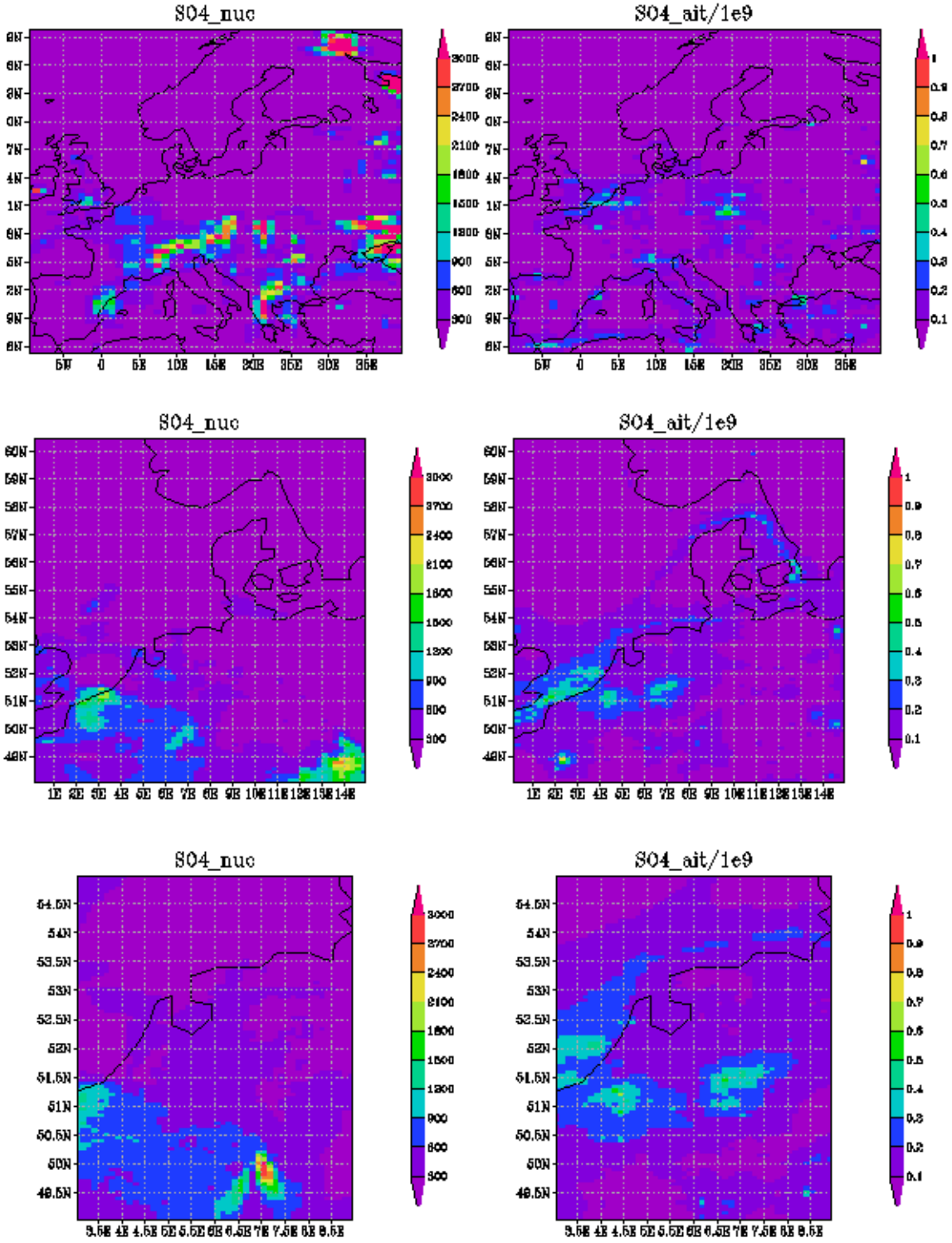

Figure 11 Sulfate concentrations of nucelation and Aitken mode (molec/cm^3), coarse resolution (upper panels), zoom factor 2 (middle) and zoom factor 4 (lower panels)

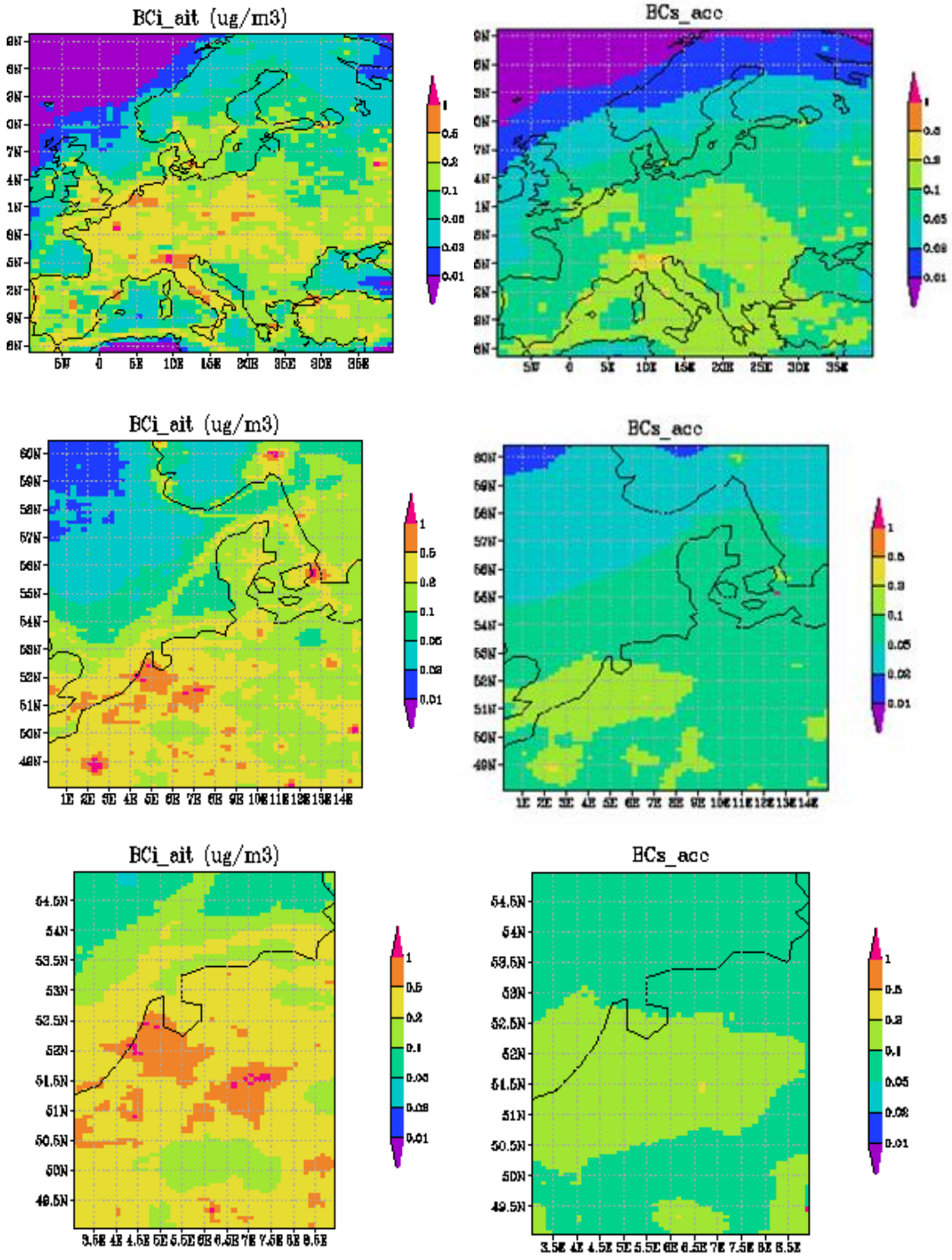

Figure 12 Annual average BC concentrations in µg/m^3 with coarse resolution (upper panels), zoom factor 2 (middle) and zoom factor 4 (lower panels)

4.3 Time series

For Cabauw, hourly mass concentrations from the Marga instrument were availaible for sodium and sulfate. These were compared with the model runs (Fig. 13,) for January (higher sea salt concentrations) and for April (more nucleation events expected). The total SO4 mass is underestimated by LOTOS-EUROS, both in the sectional and in the M7 version. This is a shortcoming that was already been found in previous model evaluations of LOTOS-EUROS. The concentrations of the two model versions and the different runs are very close to each other. Their time correlation is excellent, only the absolute value differs, with higher concentrations for the M7 model version for the moments at which high concentrations occur. It is not always the zoom factor 4 run that has the highest concentrations. Apart from the underestimation, the correlation of the runs with observations is modest, for some days it is good (days 10-12) but there are also episodes which are less well correlated (days 105-110). For sea salt the correlation is better in the sense that high concentrations are reached at the same time, but for the runs using M7 the highest concentrations are too high and vary too quickly. For sea salt, the timing of events for the sectional and the M7 version is nearly identical, since the same source function is used, but the deposition scheme is different in the two versions so that the absolute values are different. Unfortunately we do not have mass measurements for Melpitz and Vavihill for 2008. For the sectional model version of LOTOS-EUROS, it appeared that the underestimation of SO4 is a quite general model shortcoming.

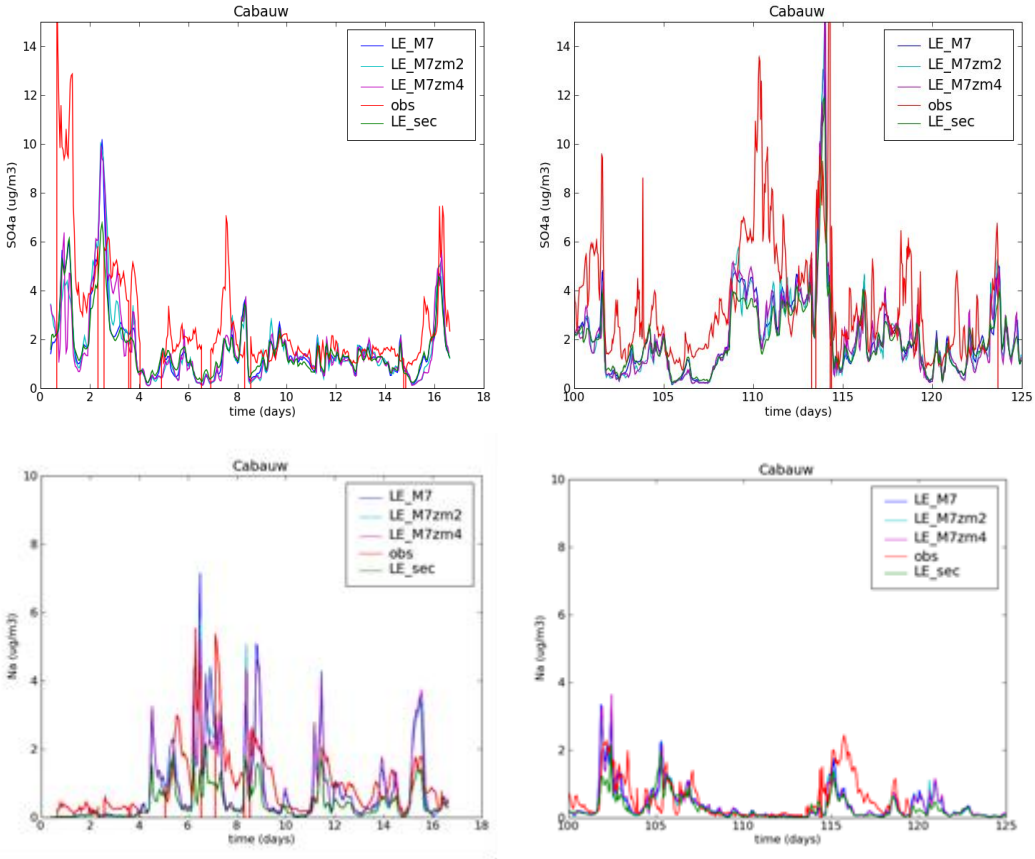

Figure 13 Sulfate and sodium mass concentrations Cabauw and model results for sectional model and for three horizontal resolutions with M7, January (left) and April (right) 2008

Particle number concentrations were observed at Cabauw with the SMPS instrument for most of the year 2008. Unfortunately, results from April and May were not available due to technical problems and servicing, and particles with a diameter smaller than 30 nm were not counted well. These observations can be translated to particle number concentrations in the size 30-100 nm and 100-500 nm, which can then be compared with the modelled Aitken and accumulation mode. There are two complications: the particles 1-30 nm and 500-1000 nm are missing in the observations, which should lead to an underestimation of the model results, but at the same time not all emission sources are present in the model, which could compensate for this. Thus, the comparison is rather qualitative. Nevertheless, figure 14 indicates that at least the order of magnitude for the two size classes is correct and that at some periods there is a general correlation between observations and model (e.g. day 42-45, 50-55, 160-165).

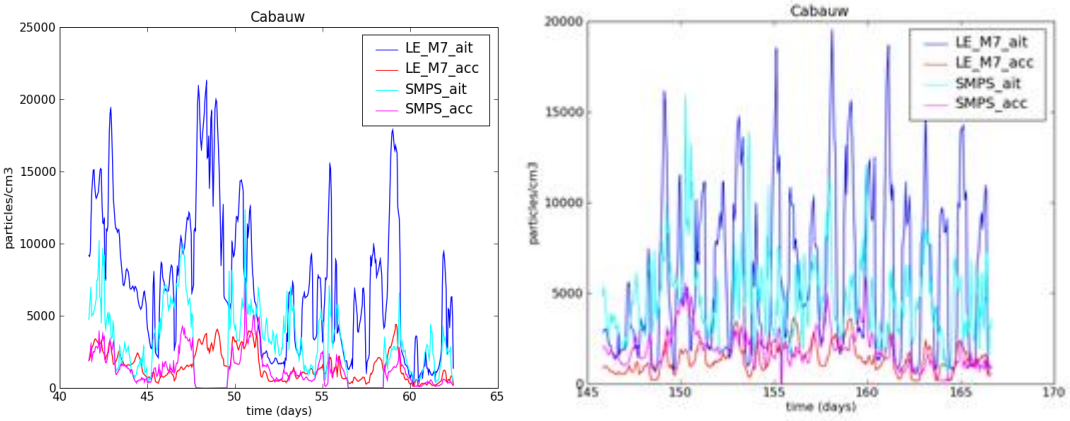

Figure 14 Observed and modelled particle number concentrations Cabauw and model, February (left) and June (right) 2008

The particle number concentrations at the different stations are nearly identical for the three model resolutions (Figs.18-21). The time correlation between the three runs is very high. There are small differences in absolute value, sometimes the zoom run has the highest concentration of a specific particle number class, sometimes the run at standard resolution. For all three stations, nucleation events are mainly found in March/April and July. This is not in full agreement with the observations of Manninen et al (2010). For Cabauw and Vavihill, they indeed find most nucleation events in spring and summer (but with a maximum in June), but more than modelled here. For Melpitz they find nucleation events nearly every day from April to August, which is not reflected by the model results.

For the coarser modes the general signature of the stations is well represented. Cabauw has the largest particle number concentrations, close to areas of high emissions. The Aitken mode, representing fresh emissions, has more particles than the accumulation mode, and receives coarse mode particles due to the influence of the sea. The Aitken and accumulation mode show some general correlation in time, also pointing at nearly emissions. In contrast to what one would expect intuitively, the concentrations are lower for the zoom runs. In this case, this can be explained

by the fact that Cabauw is close to regions with high emissions, but the grid cell containing Cabauw itself contains rather low emissions for the zoom runs. For Vavihill there are often more particles in the accumulation mode than in the Aitken mode, pointing at more aged aerosol, also the concentrations are lower than Cabauw. Due to the proximity of the sea, the coarse aerosol reaches the highest concentrations of the three stations. For this station, the effect of the zoom factor is largest of the three stations, due to the land-sea contrast which is averaged out more at standard resolution. Particle number concentrations are lowest for Melpitz, as expected. Being located far away from the sea, coarse aerosol concentrations are very low here. As expected for this rural location far away from local sources, the aerosol is more aged than for Cabauw and Vavihill, as can be concluded from the observation that the aitken mode contains often less particles than the accumulation mode. The difference between the runs with different resolution is very small.

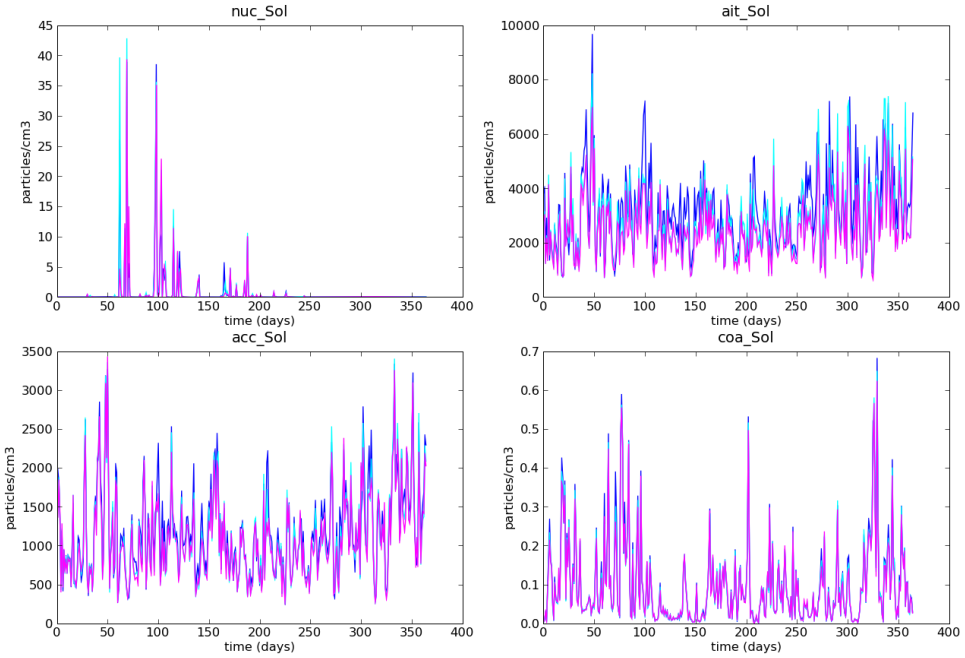

Figure 15 Daily average particle number concentrations for soluble classes, Cabauw

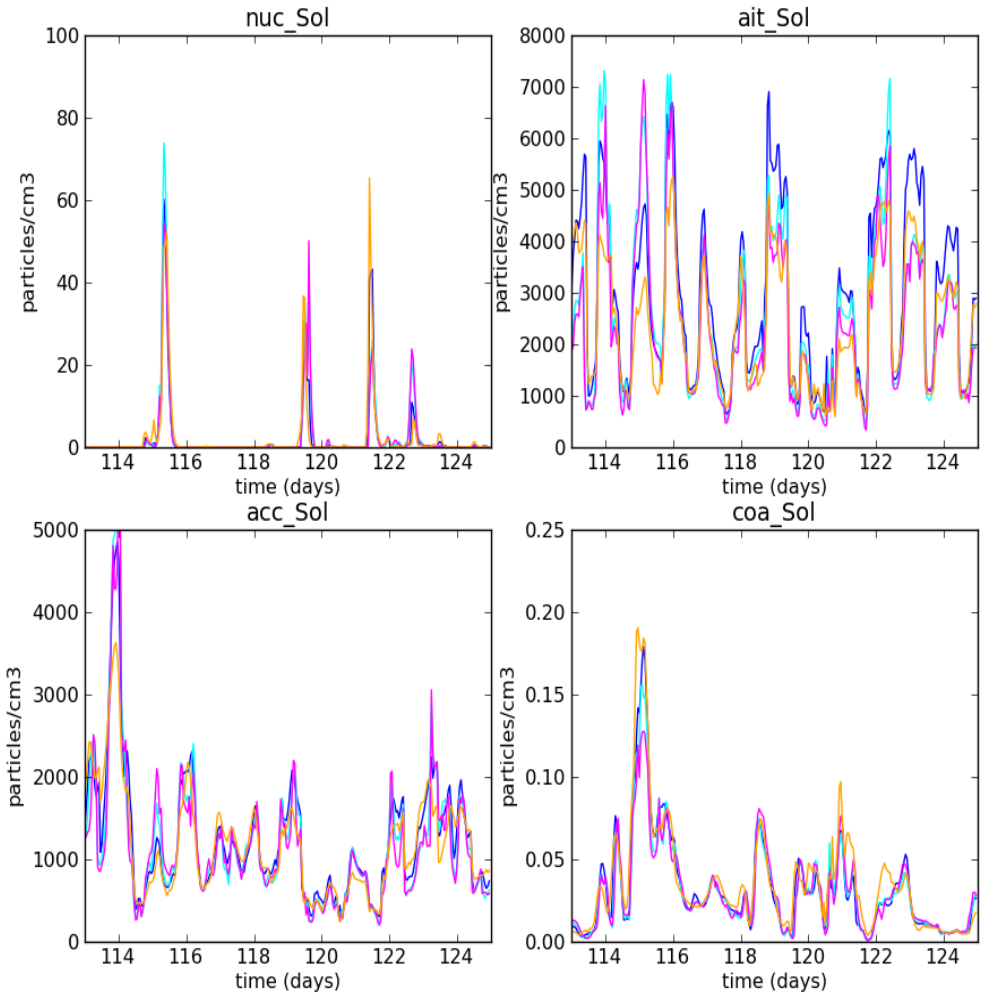

Figure 16 Hourly particle number concentrations for soluble classes, april, Cabauw. Blue: standard resolution, orange: coarse resolution, cyan: zoom factor 2, magenta: zoom factor 4

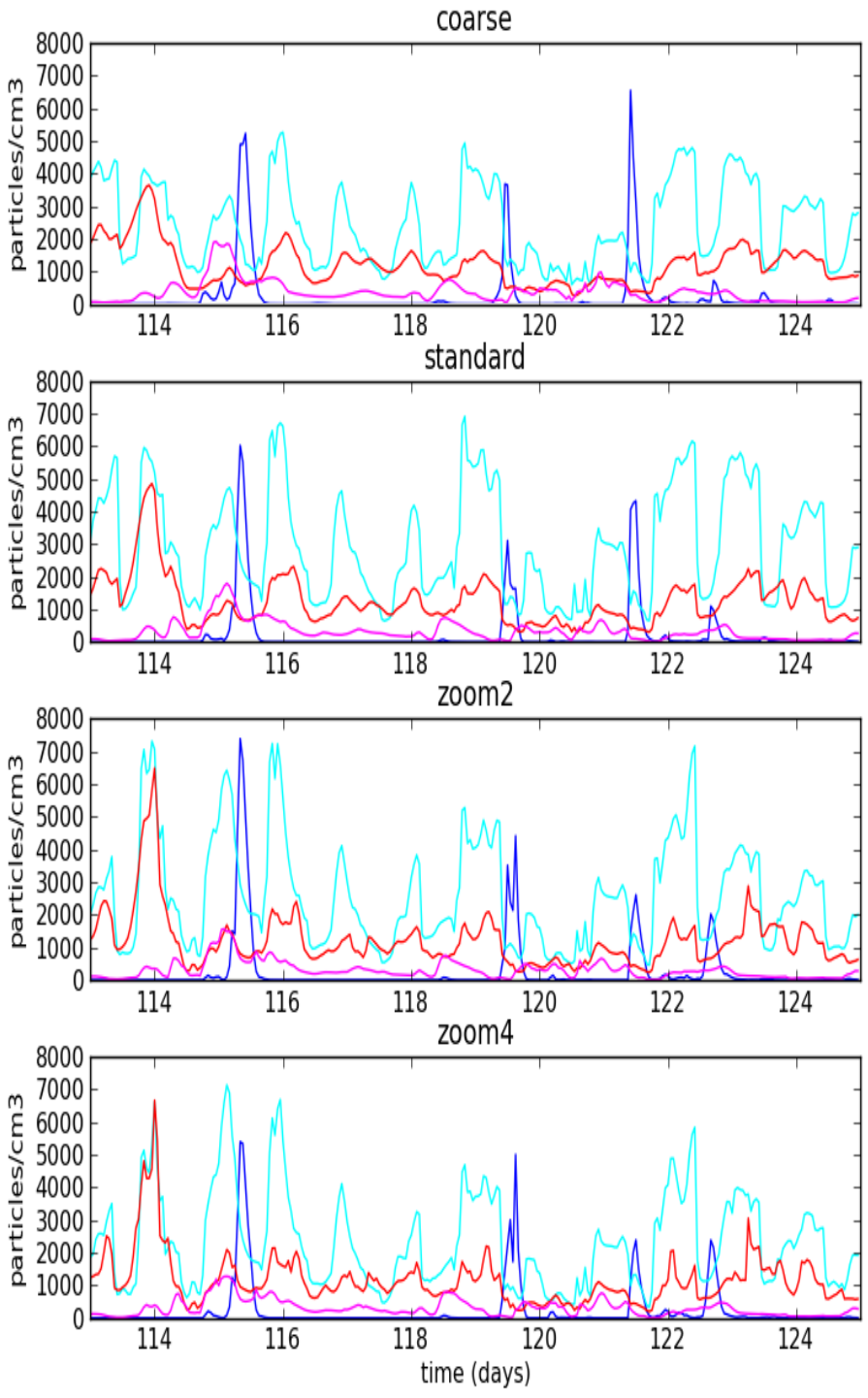

Figure 17 Particle number concentrations at Cabauw, soluble modes. Black: 100x nucleation, cyan Aitken, red accumulation, magenta 10000xcoarse

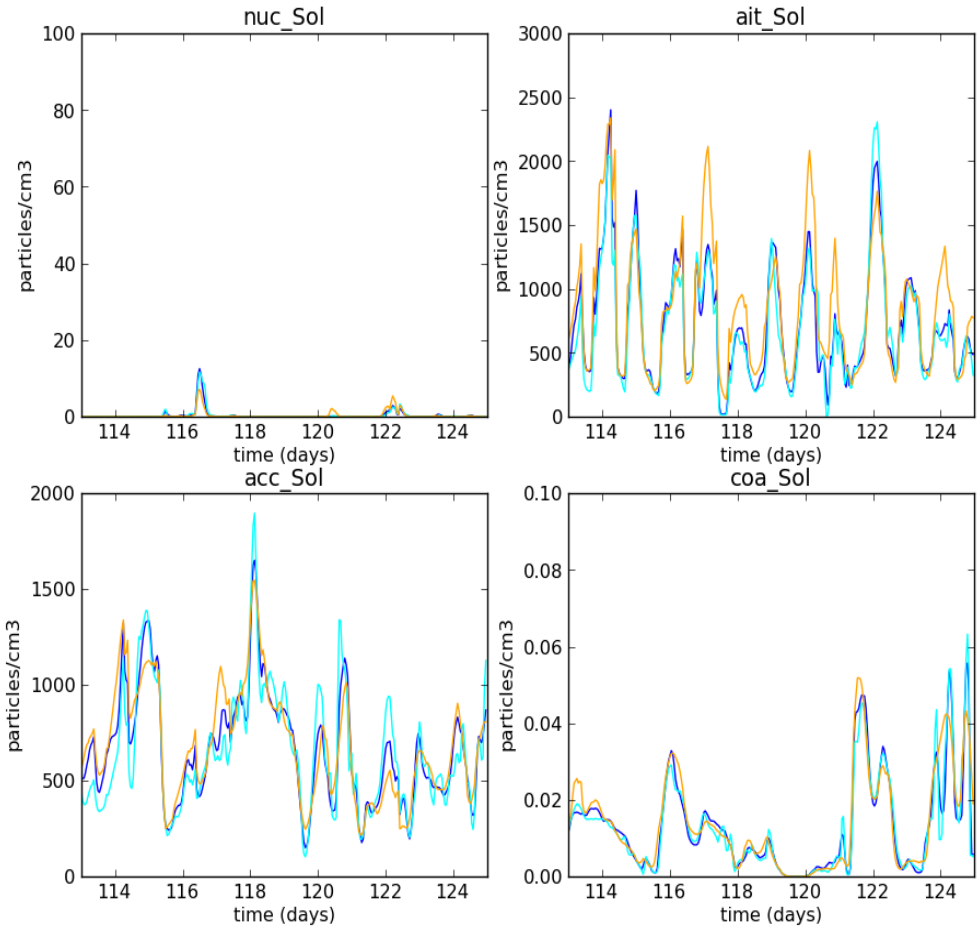

Figure 18 Particle number concentrations for soluble classes, Melpitz. Blue: standard resolution, cyan zoom factor 2, orange coarse resolution

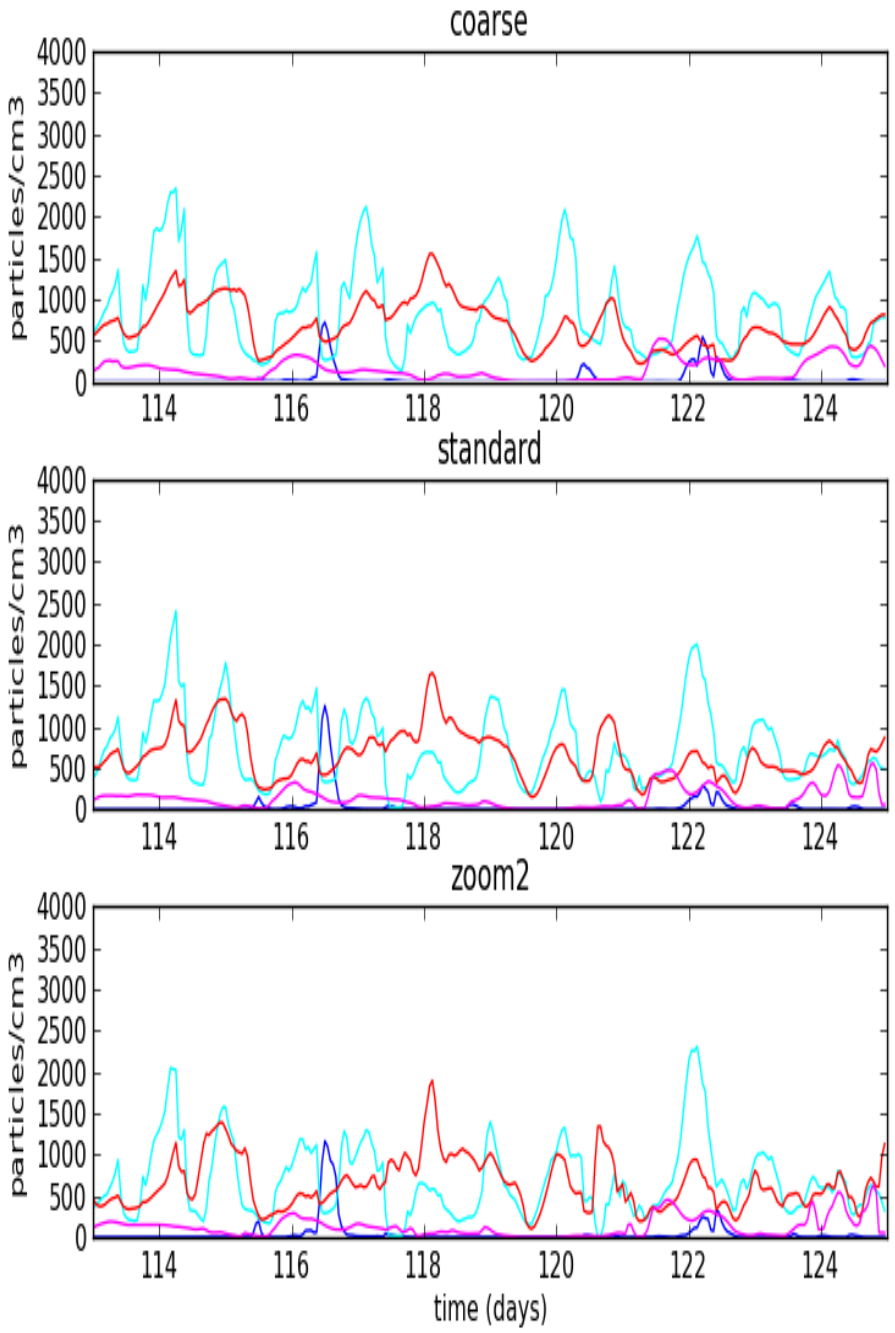

Figure 19 Particle number concentrations at Melpitz, soluble modes. Blue: 100x nucleation, cyan aitken, red accumulation, magenta 10000xcoarse

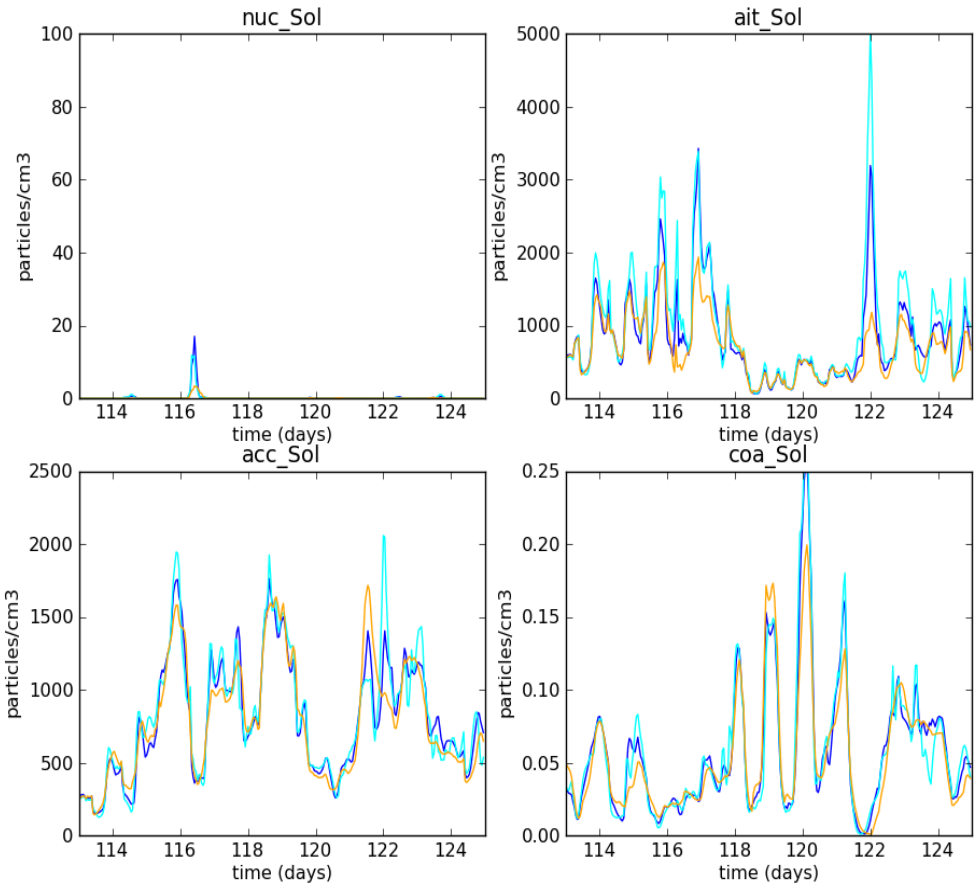

Figure 20 Particle number concentrations for soluble classes, Vavihill. Blue: standard resolution, cyan: zoom factor 2, orange: coarse resolution

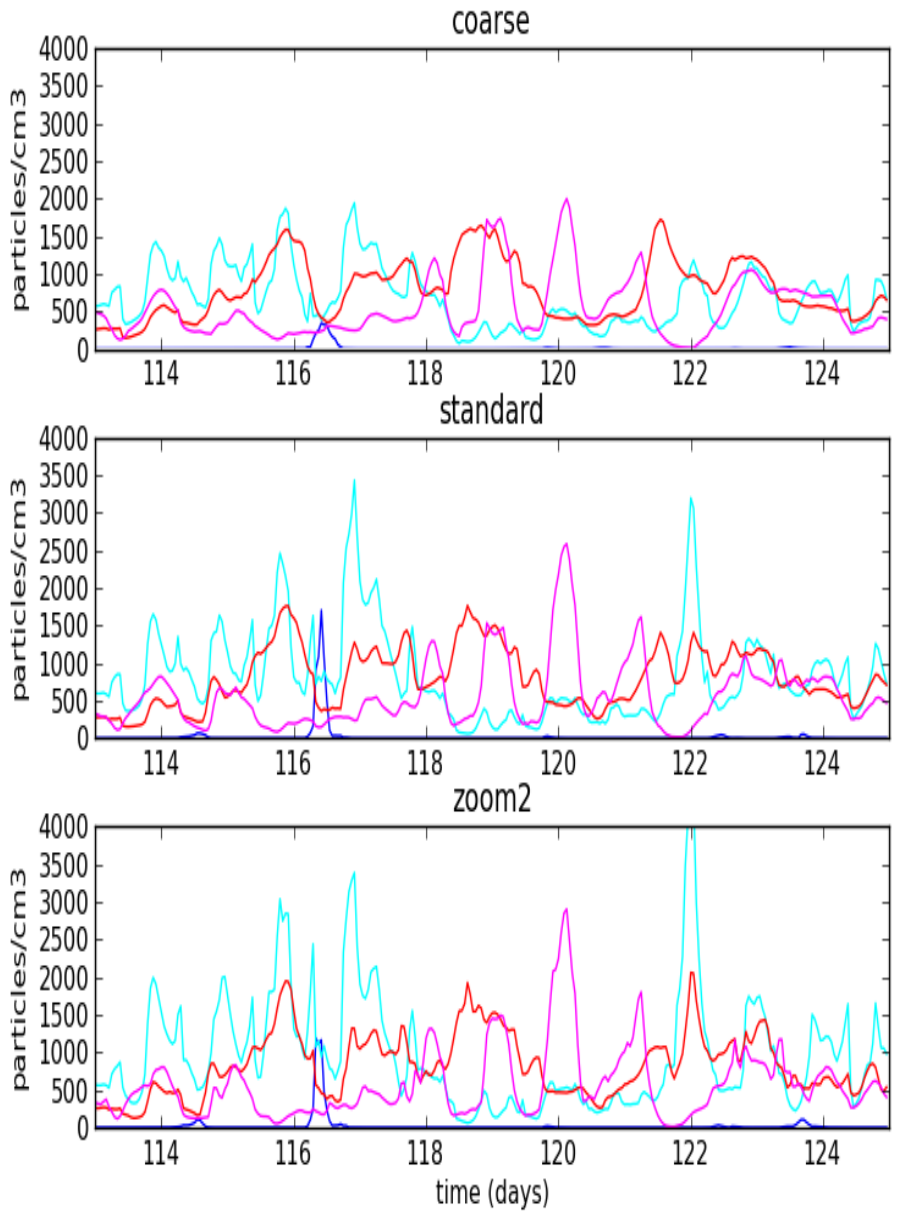

Figure 21 Particle number concentrations at Vavihill, soluble modes. Black: 100x nucleation, cyan aitken, red accumulation, magenta 10000xcoarse

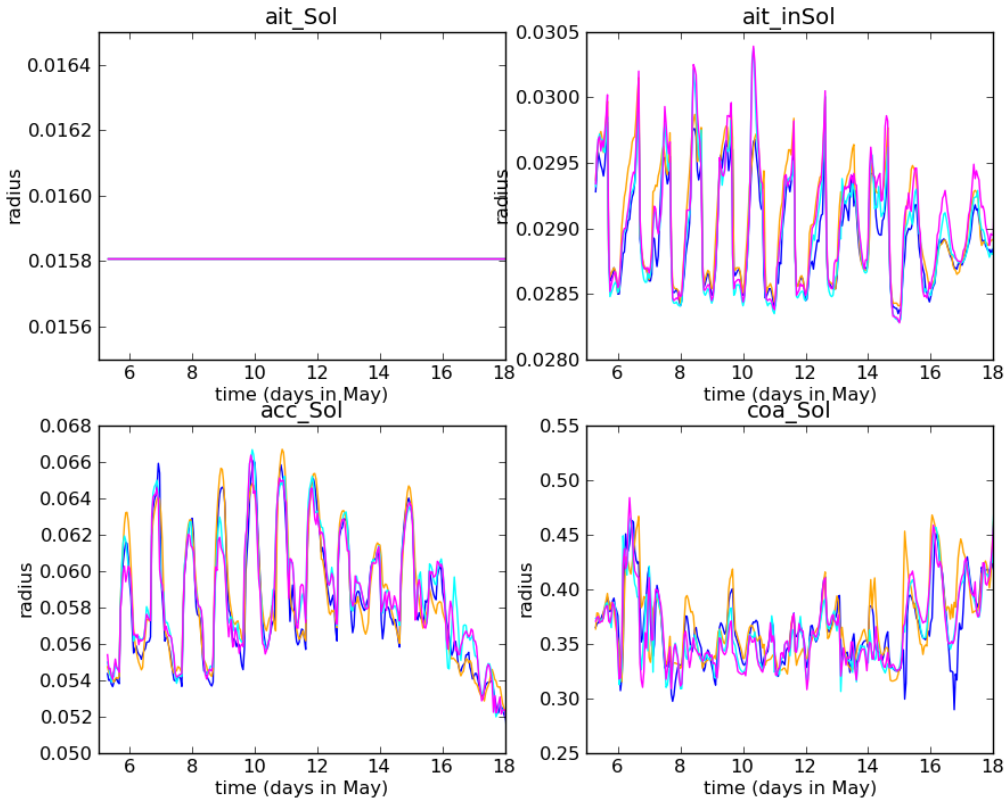

Figure 22 Modelled mass median particle radius (in um) at Cabauw, 5-18 May 2008 for the four model resolutions. Blue: standard, orange coarse resolution, cyan zoom factor 2, magenta zoom factor 4

The mass median radius of four modes at Cabauw during a warm, sunny and dry period was investigated (Fig. 22), so for an episode for which much ageing can be expected. One mainly sees the daily variability. The results for the different model resolutions show some difference, with the coarser resolutions tending to smaller radii in the Aitken mode and larger radii in the accumulation mode than the results for increase resolution. This is should be set against the time series of mass and number, where less particles in the Aitken mode and somewhat more particles in the accumulation mode were found during such episodes. The radius for the coarse aerosol is smaller than 0.5 µm for this episode with continental air arriving at Cabauw. At the end of the period, clean air from the sea comes in, which results in an increased radius for the coarse mode (fresh aerosol, less effect of deposition) and a reduced radius for the accumulation mode. Also snap shots were made to verify whether during such an episode the spatial patterns of concentrations and radius are comparable. These snapshots (Figs. 23-26) indeed indicate that gradients are sharper and more details are visible at higher resolution. Especially for the accumulation mode, there is a noteworthy difference between the highest and second highest resolution in number and dry radius (Figs 25 and 26), with more detail, differences in peak concentrations of up to 15 % and larger radii (2%) in Northwest-southeast bands for the highest resolution, so that the increased resolution has some added value. For the other modes the differences are smaller. The time series and snapshots indicate that transitions are smooth and the impact of resolution on lifetime seems therefore still small for the resolutions of LOTOS-EUROS.

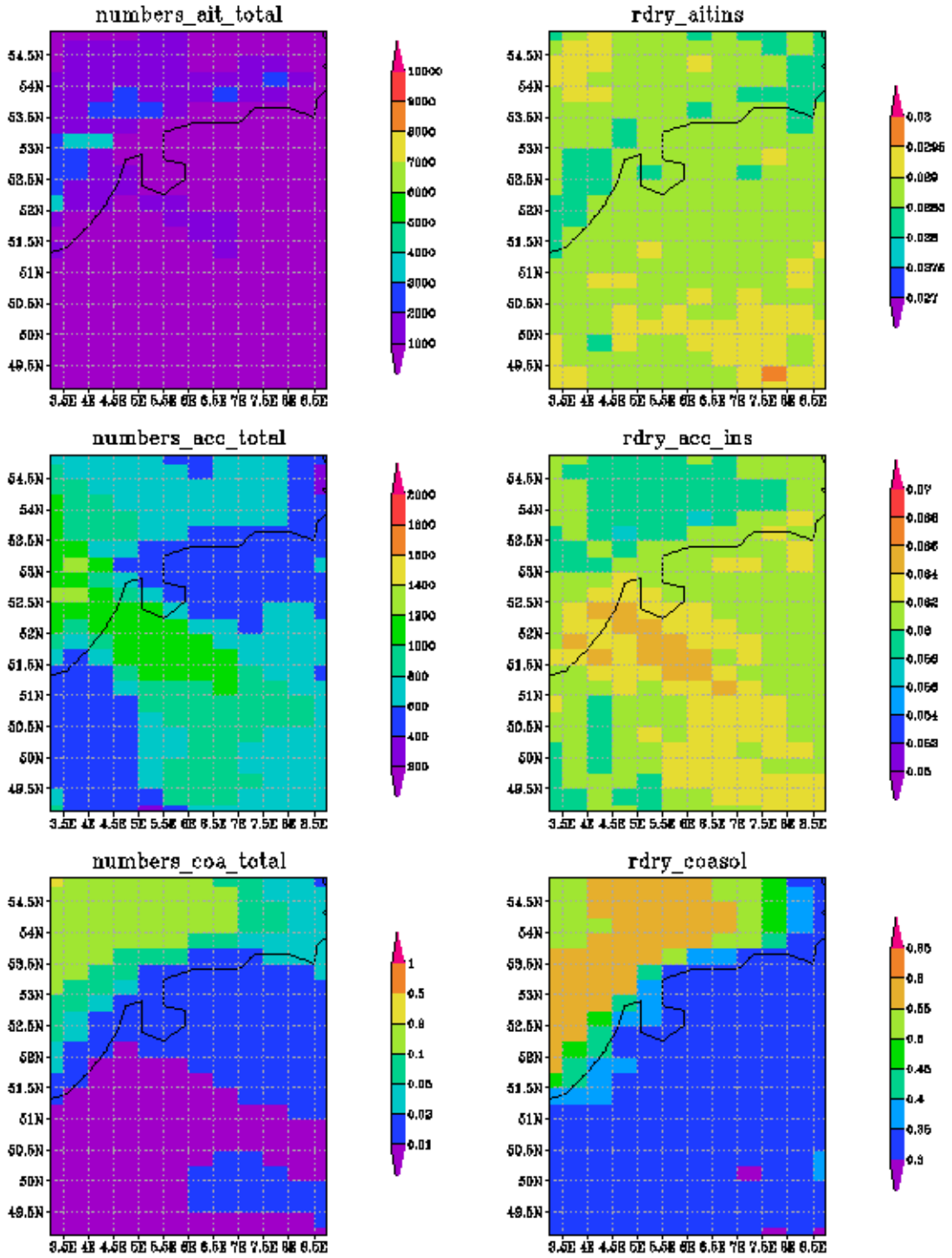

Figure 23 Snapshot of number concentration and dry particle radius, May 9 2008, 14 h, standard resolution. The dry radius ranges from 0.027-0.03 µm (Aitken), 0.05-0.07 µm (accumulation) and 0.3-0.65 µm (coarse)

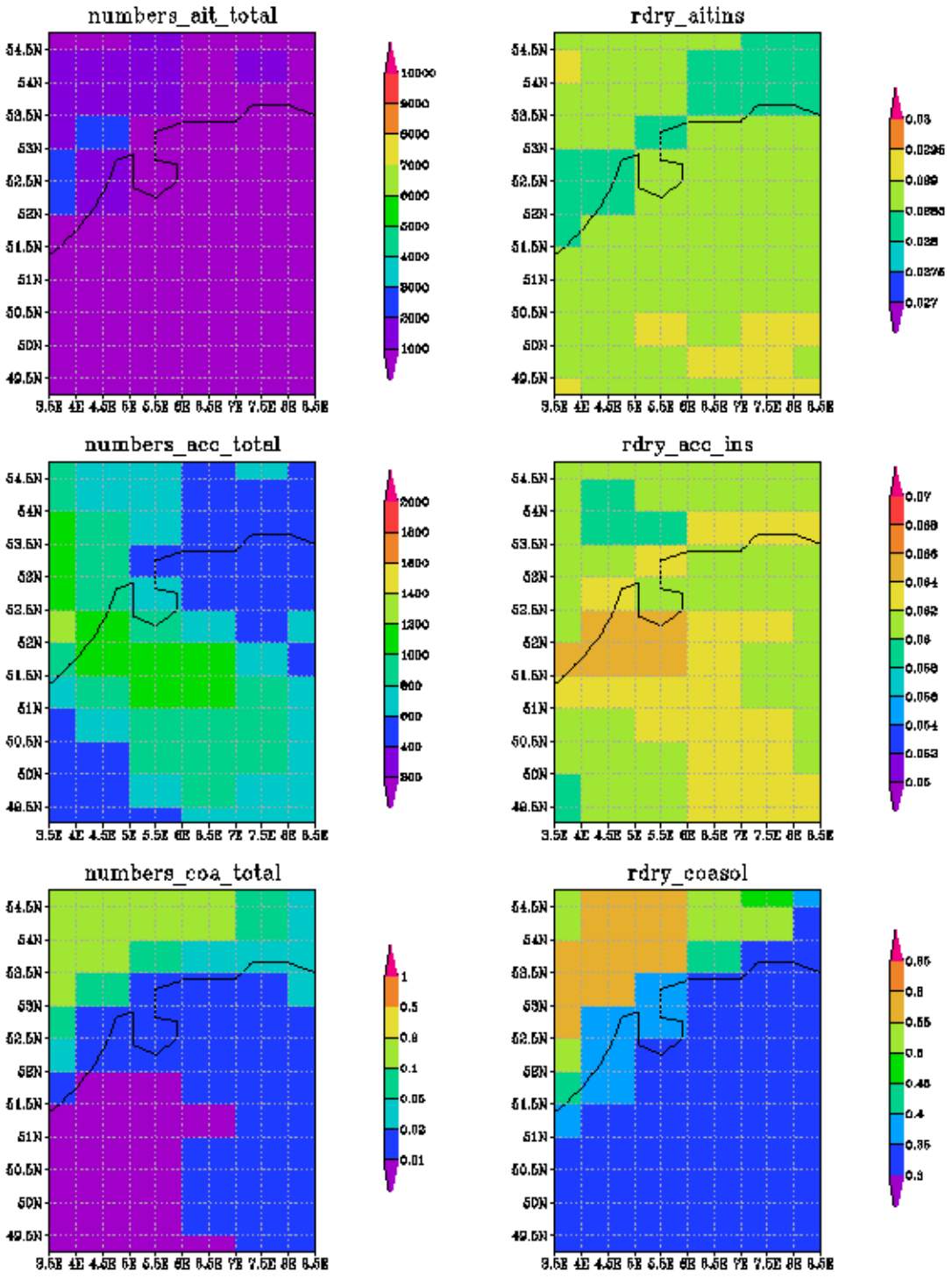

Figure 24 Snapshot of number concentration and dry particle radius, May 9 2008, 14 h, coarse resolution. The dry radius ranges from 0.027-0.03 µm (Aitken), 0.05-0.07 µm (accumulation) and 0.3-0.65 µm (coarse)

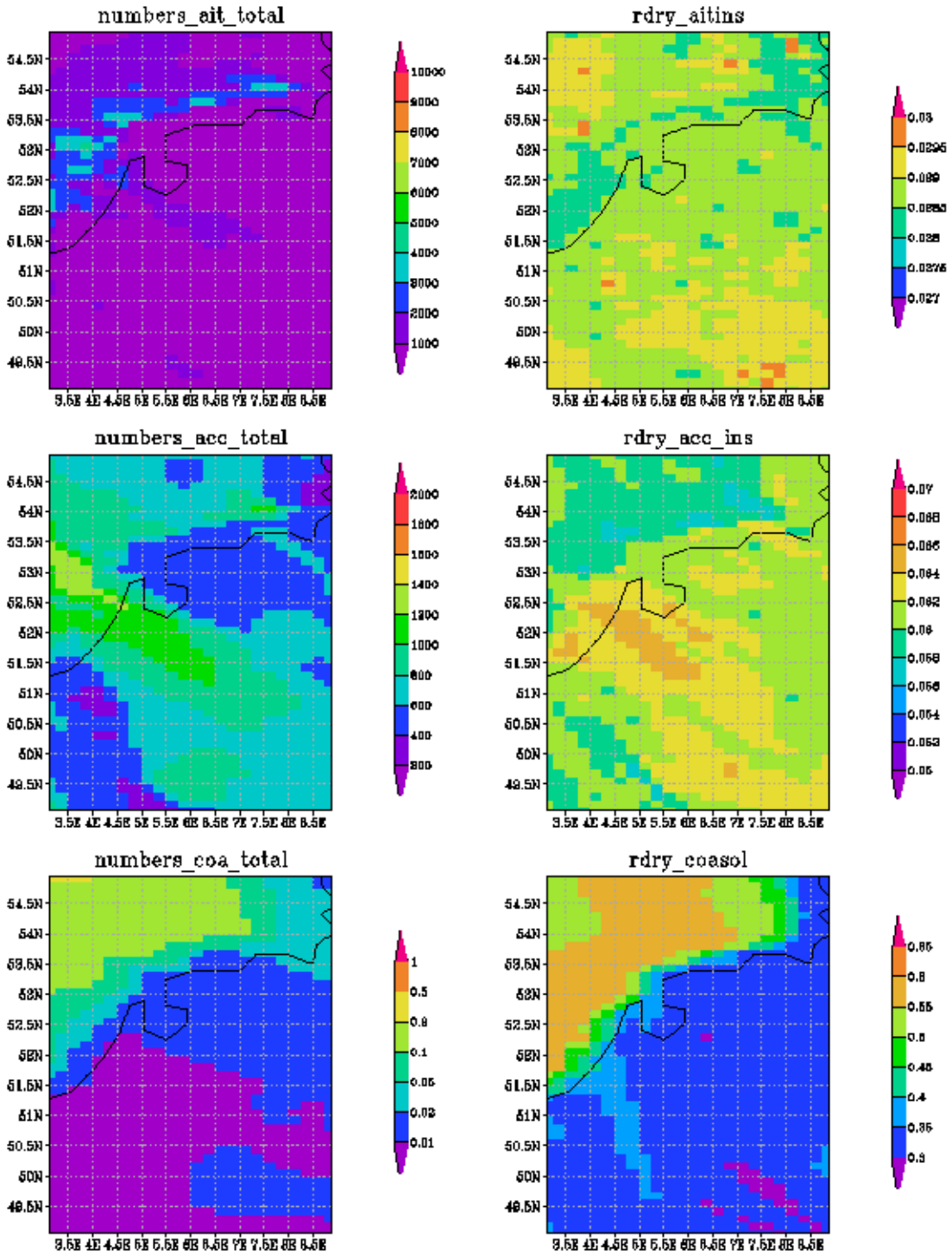

Figure 25 Snapshot of number concentration and dry particle radius, May 9 2008, 14 h, zoom factor 2. The dry radius ranges from 0.027-0.03 µm (Aitken), 0.05-0.07 µm (accumulation) and 0.3-0.65 µm (coarse)

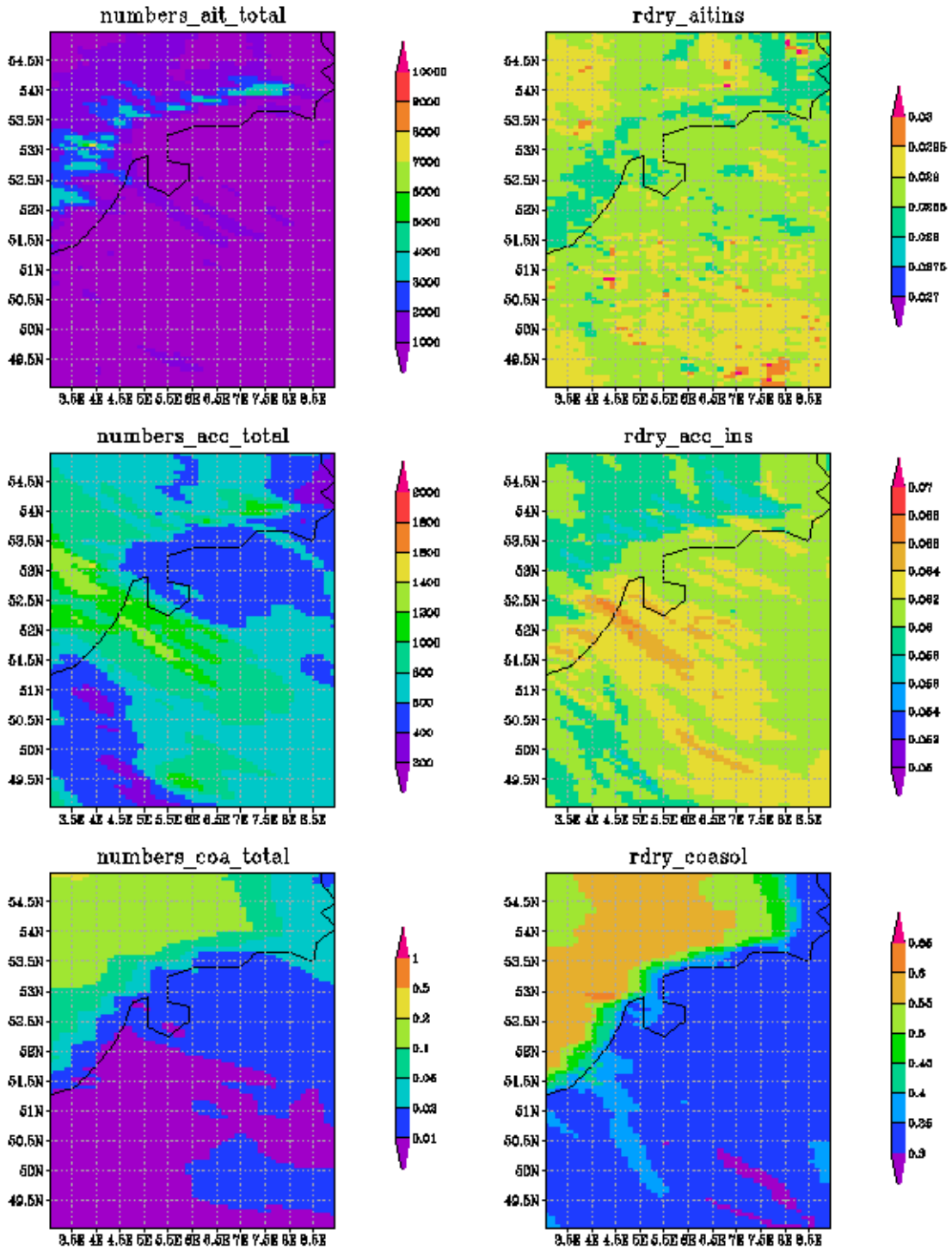

Figure 26 Snapshot of number concentration and dry particle radius, May 9 2008, 14 h, zoom factor 4. The dry radius ranges from 0.027-0.03 µm (Aitken), 0.05-0.07 µm (accumulation) and 0.3-0.65 µm (coarse)

4.4 Vertical distribution and transport

4.4.1 Vertical distribution

The three stations Cabauw, Melpitz and Vavihill have a different distribution over the vertical (Fig. 27-29). For Cabauw, high concentrations of Aitken and accumulation mode particles are found in the lower layer, whereas they are very small for the upper model layer. This is consistent with the location of the station near anthropogenic emission sources, which emit in the lowest model layers. These fresh emissions are not mixed rapidly into the higher model layers. In the highest layers, nucleation mode particles can be found with relatively high concentrations, which are closely reflected by higher concentrations of Aitken mode particles. High concentrations of nucleation mode particles at ground level seem related to high concentrations at high levels, but with some hours delay. This could be due to vertical mixing, which increases during the morning, but also due to a difference in wind velocity, with stronger winds at higher level. For Melpitz, the difference between the layers is much smaller, being away from direct sources the particles are more evenly distributed in the vertical, with lower accumulation mode particles near the ground and slightly higher concentrations of accumulation mode particles at the highest level, as compared to Cabauw. For Vavihill (at standard resolution) the picture is comparable to that of Melpitz. For these two stations, nucleation mode particle concentrations were so low that they can hardly be observed from the present scale. Also indicated in Figs 22-24 is the classification of nucleation events (Manninen et al 2010). The events are observed at low level but are plotted here at the highest level since in the model nucleation mode particle concentrations are highest there. An event is classified as a yes, no or undetermined, positive identifications are plotted as a cross at 1000 and is assigned to midday, regardless of the absolute timing. The correspondence between modelled and observed events is rather poor: many events are missed, in particular for Melpitz.

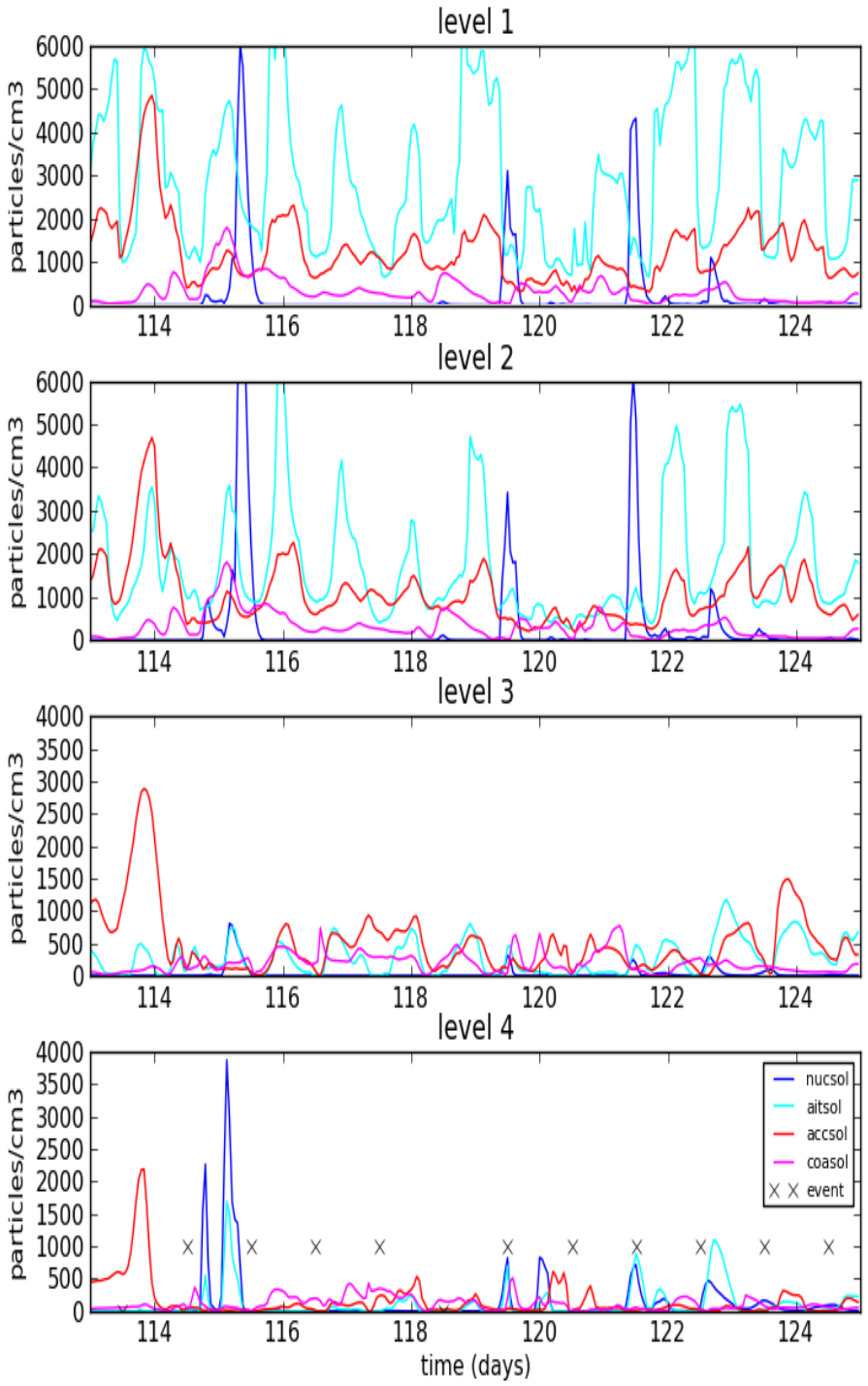

Figure 27 Cabauw. Particle number concentrations and observed event identification. At level1 and 2, the nucleation mode concentration has been mulitplied by 100, the coarse mode has been mulitplied by 10000 at all levels

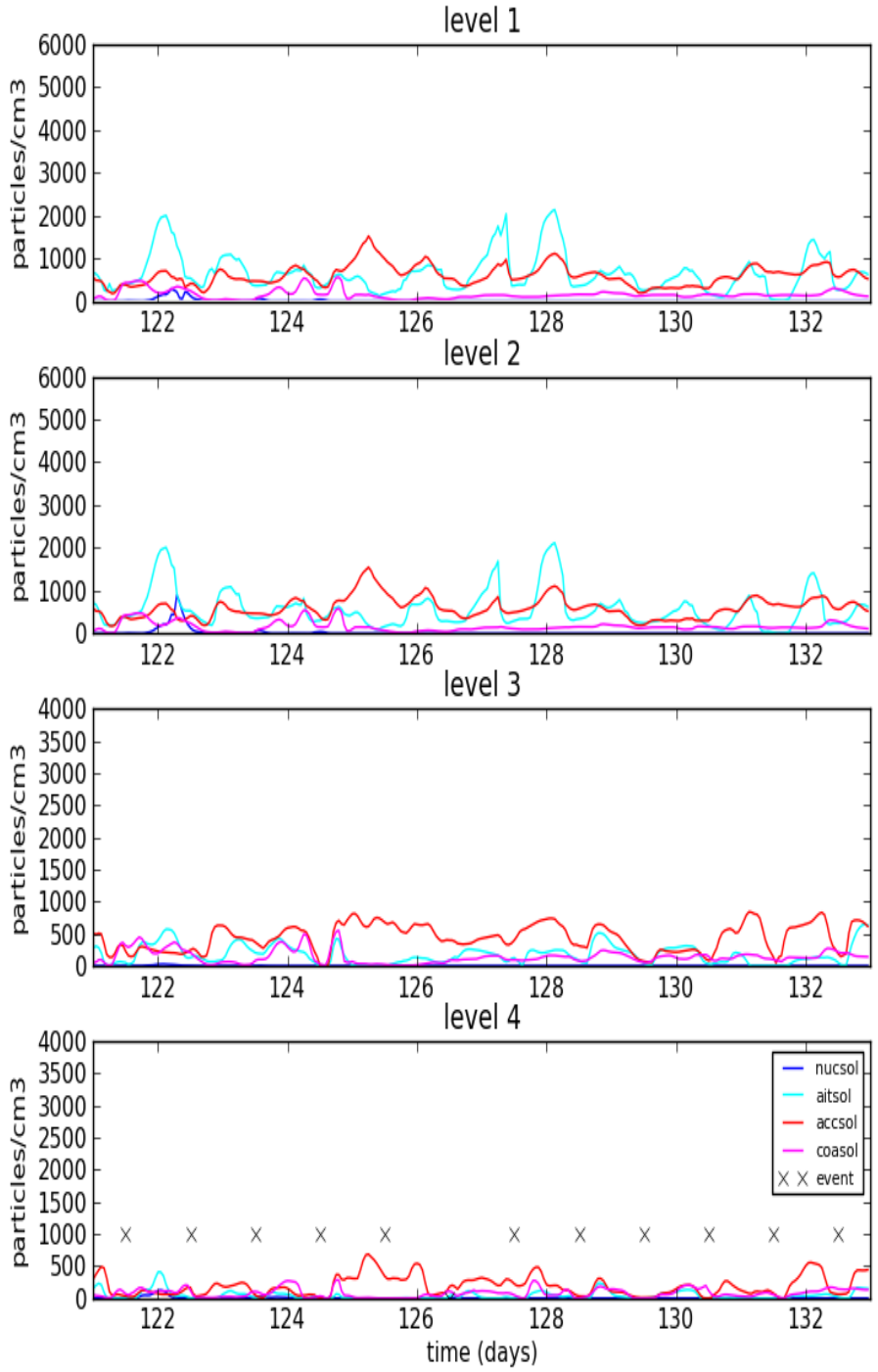

Figure 28 Melpitz Particle number concentrations and observed event identification. At level1 and 2, the nucleation mode concentration has been mulitplied by 100, the coarse mode has been mulitplied by 10000 at all levels

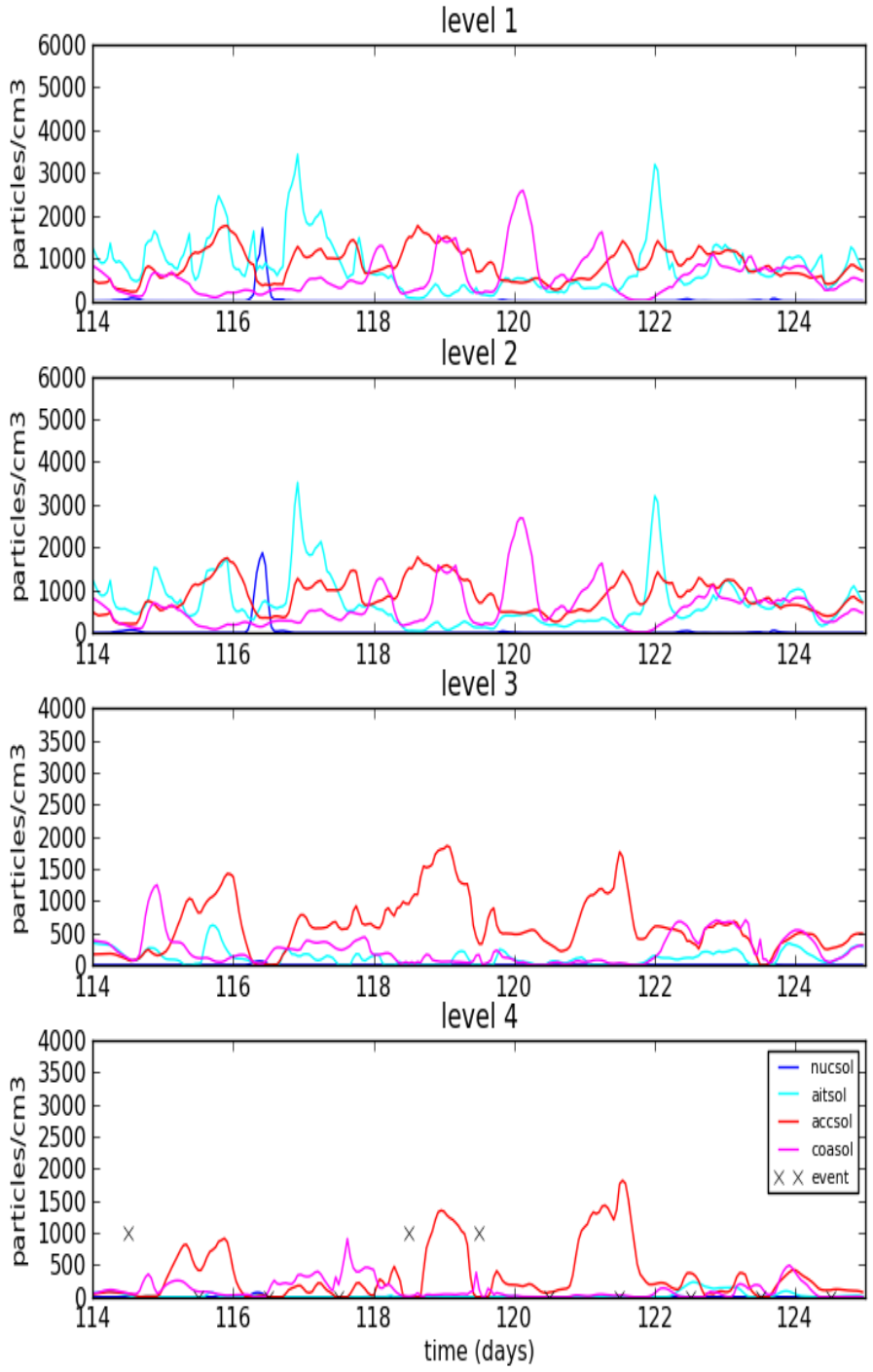

Figure 29 Vavihill. Particle number concentrations and observed event identification. At level1 and 2, the nucleation mode concentration has been mulitplied by 100, the coarse mode has been mulitplied by 10000 at all levels

4.4.2 Origin and transport of nucleation mode particles

Hovmoeller plots were made at several latitude and longitude sections to illustrate transport and eventually the origin of nucleation mode particles. The time section is from day 100-day 125. The latitude cross-sections (Figs. 30 and 31) confirm the finding that nucleation mode particle concentrations are considerably higher at the highest model level. West of 15 E, transport is mostly eastward, so from the ocean/sea/coastal areas inland. Most events can be followed for about 24-48 hours, but the first event seems to reach as far as 25 E, taking nearly 4 days. Events starting in the east tend to be transported westwards. At 52 N, it can be observed that the eastward propagating events do reach Cabauw, but not always Melpitz. On days with little transport, the patterns extend over 5-10 latitude and seem to have a day-night pattern. The events identified in the time series can also be found in the Hovmoeller plots. The longitudinal sections show less transport than the latitudinal sections. This indicates that events occur over latitudinal areas of 10 degrees or even larger and are transported westward or eastward, with most of the nucleation events originating from the sea (clean air). To explain the observed nucleation events in Melpitz, which are more abundant than in Cabauw, other nucleation mechanisms should be included in the model.

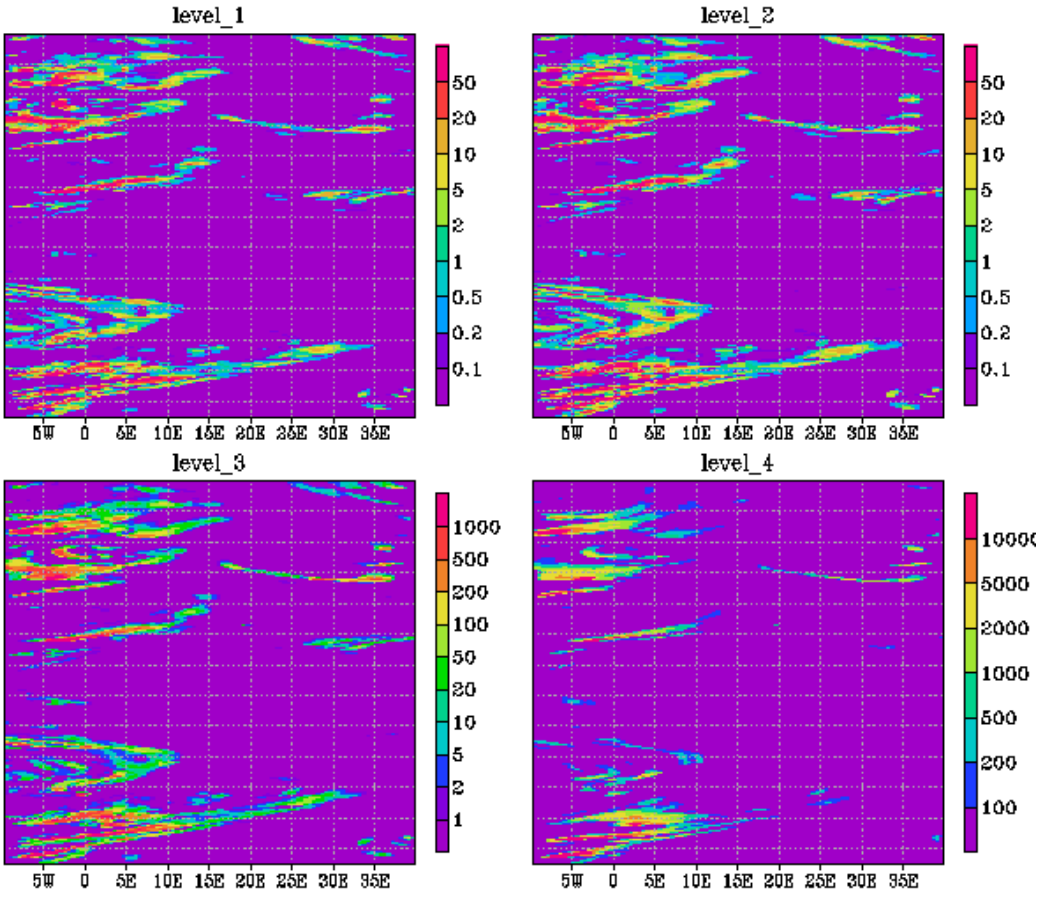

Figure 30 Hovmoeller of nucleation mode (particles/cm3) plot at 52 N(Cabauw, Melpitz), t=9 April-4 May. Horizontal axis: longitude, vertical axis time (ticks are 2 days). Note the difference in scaling for the different model levels

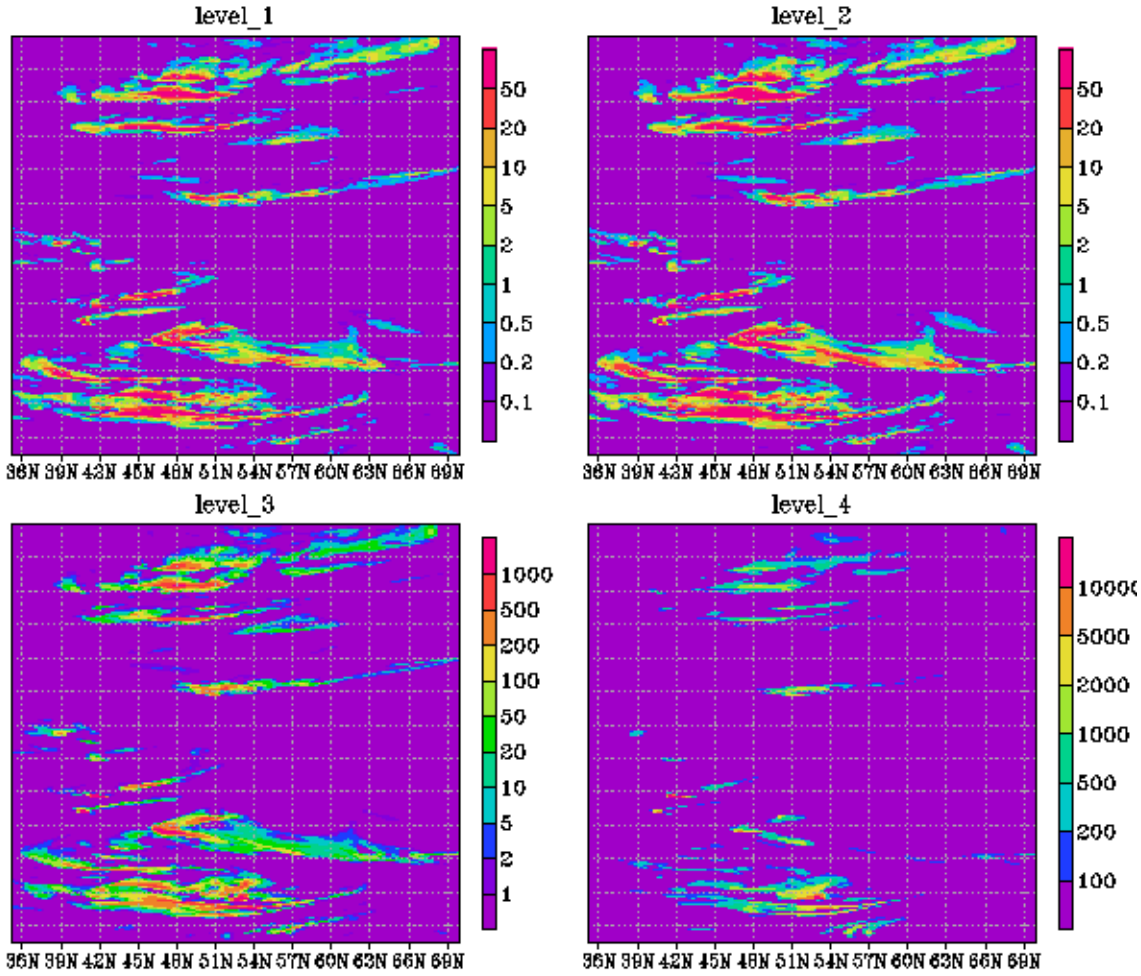

Figure 31 Hovmoeller plot of nucleation mode particles (particles/cm3), 5˚ E (Cabauw), t= 9 April-4 May. Horizontal axis: latitude, vertical axis time (ticks are 2 days). Note the difference in scaling for the different model levels

5 Discussion and recommendations

The correspondence between the sectional version of LOTOS-EUROS and the version with M7 indicates that M7 module was implemented successfully. The time correlation between the two model versions is excellent, with slightly higher sulfate and sea salt concentrations for periods of very high concentrations. Since the sources are identical in the model versions, this must be the effect of changes in the size distribution and the different deposition scheme. Differences are largest for the coarse sea salt aerosol, for which the difference in deposition velocity is particularly large as already found in a previous study (Schaap et al 2009). LOTOS-EUROS with M7 reproduces the general features of the observations at Cabauw quite well. Sulfate concentrations are underestimated, which is a general feature of LOTOS-EUROS, also in the sectional version, but the time correlation is quite good. Also particle number concentrations, which could be compared with SMPS observations, showed good correspondence with number concentrations in terms of absolute concentrations and variability and for some periods showed model and measurements showed a good correlation. This is despite the fact that some sources are missing in the model or are incorporated in a simplified way and the fact that the observed particle number size classes do not fully the size range of the model size classes.

The main question was whether the resolution would have an impact on aerosol dynamics. For the annual averages, the runs with horizontal resolution did not show large differences with the run at standard resolution. In emission hotspots, concentrations were slightly higher due to less direct dilution, and more horizontal detail was found, especially for the accumulation mode on a warm and sunny day, but there seemed no fundamental difference in lifetime of species. Annual average concentrations of black carbon, sulfate and sea salt compare well with other studies, e.g. for Europe using TM5 (Aan de Brugh et al 2010) for 2006 at even coarser resolution (1x1º) and using a different emission and land use database.

The time resolution of the model is not an issue in the present study. Particles were observed to grow rapidly (Manninen et al 2010) and may quickly (within one hour) grow from the nucleation mode into the Aitken and even the accumulation mode. Box experiments with M7 (Vignati et al 2004, repeated but not shown in the present study) showed that for the nucleation mode, particle number concentrations indeed may change by three orders of magnitude or more within 3 hours. This is in agreement with the observed behaviour. For the larger modes, the timescales were slower. The time steps in LOTOS-EUROS were compatible with the M7 time steps.

The typical signatures of the three stations Cabauw, Vavihill and Melpitz were represented correctly. For an increased resolution, there were minor differences. For Cabauw, concentrations were slightly lower than for the standard resolution, for Vavihill, the differences are larger, which is an effect of the proximity of the sea, so that the grid cell at standard resolution contains more sea surface and is therefore less representative for the station. Also when looking at snapshots and the development of the dry particle radius some differences were observed but they were small.

Although the mass distribution and the Aitken and accumulation mode are modelled quite well, the number of modelled nucleation events does not match observations. Relatively few nucleation events were found, in particular for Melpitz for which many nucleation events were observed. Nucleation events take typically 3 hours in field observations. Since air is transported and the observations are fixed in space, this indicates that these events take place over a rather wide area. For very low wind speeds, say 2 m/s, this means that their spatial scale is about 22 km, which is the size of a grid cell in standard resolution at a latitude around 50 N, and it is very likely that the spatial scale of the event is larger. In the model, high concentrations of nucleation mode particles are only found higher up in the atmosphere and have their origin mostly above sea of ocean and then seem to propagate eastwards. They are related in time with (much lower) concentrations of nucleation mode particles lower in the atmosphere. These concentrations may reach lower levels by vertical exchange, which would explain small time differences between higher concentrations aloft and near the surface. But another explanation for the modelled concentrations would be that the nucleation event occurs in the whole column at once but is transported more quickly aloft than near the ground, due to higher wind speeds there. Most models underestimate the nucleation mode particle concentrations. Explanations for this are an overestimation of the condensation sinks, and underestimation of the nucleation rate or the contribution of other nucleation pathways that are not accounted for in M7, as already suggested by Stier et al (2005).

The present model set-up is probably good enough to have answered the main question. Still, the model can be improved on the following points, partly technical, partly more fundamental. In order of relevance, these points are:

- Use a better emission database with more info on particle size and number. Such an emisson inventory has become available as part of the EUCAARI project (Denier van der Gon et al 2010).
- Translate the default boundary conditions to the M7 species or use boundary conditions from a global model (e.g. TM5)
- Improve the coupling between the species that are used in the chemistry of LOTOS-EUROS and the species used in M7. At present, there is a one-way coupling but no feedback.
- Improve emission parameterization for small sea salt particles (Martensson instead of Monahan)
- Take dust emissions into account
- Take nitrate/ammonium into account
- Take organic aerosols into account
- Include wildfire emissions
- In vertical: use the (height-dependent) atmospheric pressure for physical processes, now ground level pressure was used at all levels
- Take in-cloud scavenging into account (general improvement of LOTOS-EUROS)
- Sedimentation was applied for all modes, although it is only relevant for the accumulation and coarse modes (speed-up)
- In the sectional approach of LOTOS-EUROS, all H2SO4 is directly passed to the SO4 aerosol phase. Therefore, deposition of H2SO4 is not taken into account. This is probably of minor importance, since the transfer of high concentrations of H2SO4 to SO4 is a rather fast process in M7.

Nevertheless, low concentrations of H2SO4 can be persistent in LOTOS-EUROS with M7

- Include other nucleation mechanisms than binary nucleation, experiments with nucleation rate (improvement of M7 itself)

For a more thorough study of events, more frequent model output than hourly or precise hourly budgets may be needed to study the contribution of the different aerosol processes in more detail. The results should be compared with detailed observations that are available from different monitoring sites. Ideally, the model should contain the improvements mentioned above.

6 Conclusions

The aerosol dynamics module M7 was coupled to LOTOS-EUROS successfully. Modelled mass concentrations were well correlated in space and time with the sectional version of LOTOS-EUROS. Runs with increased or reduced resolution did have an impact on the level of spatial detail (stronger gradients, higher concentrations in urban or industrial areas for higher resolution). The impact of resolution on lifetime of species was not very clear. Time series at different resolutions were well correlated, with nearly identical timing of events but with the obvious effect of dilution, with a few exceptions that may point at lifetime effects. This indicates that at the spatial scales varying from 1/8x1/16 to 1x1/2º, the resolution seems not a crucial factor, although on the smallest scale on a warm and sunny day more detail was visible and the radius of the particles was increased slightly.

However, it is expected that near sources (at street level, in plumes) and in complex terrain (urban environment) processes at small temporal and spatial scale will become important, both due to higher concentrations and micrometeorology. These scales cannot be resolved with a model like LOTOS-EUROS and can partially be accounted for by a smart translation of an emission inventory.

LOTOS-EUROS with M7 can and should also be improved on other issues to fully exploit the potential of the model. The model should be compared thoroughly with more observations and other models to further investigate its weaknesses and strengths.

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8 Signature

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-

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A Technical details

The basic version is v1.5 with patch 026.maori and projects bestguess and LEM7_new. In the rc-file, there is a new option in the chemistry section:

```
! m7 aerosol dynamics s needed (yes,no) ?
m7_aerosol : yes
```

The chemistry for gases should be switched on (chemistry mode CBM4) to produce H2SO4 gas for condensation. Other processes of the default LOTOS-EUROS model (primary aerosols, chemistry for secondary aerosol, sea salt and dust emissions) are detached completely in the present version and can be neglected (set options to `no`).

The output of the M7 variables can be handled analogous to the concentration and meteorological fields. Variable names are defined in table A1.

Table A1 Names of indices and variables. SO4a: sulfate, BC: black carbon, OC: organic carbon, SS: sea salt, DU: dust, #:number of particles (part)

description	LE variable/index name	output variable name	unit
concentration variables	c(:,:,:,index)		
H2SO4 gas phase	i_so4g	h2so4g	#molec/cm3
SO4a nucleation	i_so4ns	so4nuc	#molec/cm3
SO4a aitken	i_so4ks	so4ait	#molec/cm3
SO4a accumulation	i_so4as	so4acc	#molec/cm3
SO4a coarse	i_so4cs	so4coa	#molec/cm3
BC insoluble aitken	i_bcki	bciait	µg/m3
BC soluble aitken	i_bcks	bcsait	µg/m3
BC soluble accumulation	i_bcas	bcsacc	µg/m3
BC soluble coarse	i_bccs	bcscoa	µg/m3
OC insoluble aitken	i_ocki	ociait	µg/m3
OC soluble aitken	i_ocks	ocsait	µg/m3
OC soluble accumulation	i_ocas	ocsacc	µg/m3
OC soluble coarse	i_occs	ocscoa	µg/m3
SS soluble accumulation	i_ssas	ssacs	µg/m3
SS soluble coarse	i_sscs	sscos	µg/m3
DU soluble accumulation	i_duas	duacs	µg/m3
DU soluble coarse	i_ducs	ducos	µg/m3
DU insoluble accumulation	i_duai	duaci	µg/m3
DU insoluble coarse	i_duci	ducoi	µg/m3
# soluble nucleation	i_nnus	nucsol	#part/cm3
# soluble aitken	i_nais	aitsol	#part/cm3
# soluble accumulation	i_nacs	accsol	#part/cm3
# soluble coarse	i_ncos	coasol	#part/cm3
# insoluble aitken	i_naii	aitins	#part/cm3
# insoluble accumulation	i_ncos	accins	#part/cm3
# insoluble coarse	i_ncoi	coains	#part/cm3

other variables (output in xxx_meteo-files)			
particle radius soluble nucleation	rdrym7modes(:,:,:,1)	rdnucs	cm
particle radius soluble aitken	rdrym7modes(:,:,:,2)	rdaits	cm
particle radius soluble accumulation	rdrym7modes(:,:,:,3)	rdaccs	cm
particle radius soluble coarse	rdrym7modes(:,:,:,4)	rdcoas	cm
particle radius insoluble aitken	rdrym7modes(:,:,:,5)	rdaoti	cm
particle radius insoluble accumulation	rdrym7modes(:,:,:,6)	rdacci	cm
particle radius insoluble coarse	rdrym7modes(:,:,:,7)	rdcoai	cm