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Determination of the Optimum MM5 Configuration for Long Term CMAQ Simulations of Aerosol Bound Pollutants in Europe

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Abstract Realistic meteorological fields are a prerequisite for the determination of pollutant concentrations and depositions by means of a chemistry transport model. Different configurations of the 5th generation NCAR/Penn State University mesoscale meteorological model MM5 were tested to determine the optimum set up for long term hindcasts that cover several months up to years. Four dimensional data assimilation (FDDA) significantly enhances the spatio temporal representation of temperature, humidity and wind. Best agreement with radiosonde observations could be achieved when temperature, humidity and wind were grid nudged every 6 hours. The quality of the resulting meteorological fields showed no significant systematic temporal or spatial variation over Europe in a model run of the year 2000. It was found that the hydrological cycle was not correctly reproduced by the model when no nudging was applied. The relevant model run showed too high relative humidity and too high rainfall when compared to observations. This led to considerably lower aerosol con-

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centrations close to ground and a shift in the deposition patterns of particle bound pollutants like the carcinogenic benzo(a)pyrene (B(a)P).

Keywords Chemistry transport model · Mesoscale meteorological model · Four Dimensional Data Assimilation (FDDA) · Aerosol · Benzo(a)pyrene

1 Introduction

Three dimensional Eulerian chemistry transport models (CTMs) like the Community Multiscale Air Quality (CMAQ) modeling system need realistic meteorological fields as input data. These fields have a significant impact on the results of the CTMs and they contribute to uncertainties in simulations of the atmospheric distribution of chemical species and aerosols. Therefore, the assessment of the quality of meteorological simulations used for chemistry transport studies before they are applied is indispensable.
At GKSS, the 5^{th} generation NCAR/Penn State University mesoscale meteorological model (MM5) [12] is used as meteorological preprocessor for CMAQ. MM5 can be used in a variety of configurations. This concerns primarily the different parameterisations of the subscale processes like the representation of cumulus clouds, the cloud microphysics, the representation of the planetary boundary layer (PBL) and the radiation. The model can be run with or without a land surface module and in different vertical resolutions. Furthermore, for hindcast studies four dimensional data assimilation (FDDA) techniques can be applied to force the simulated fields to stay close to the observations. Stauffer and Seaman [21] investigated the continuous nudging type where forcing functions are added during the entire model run. These forcing functions can be derived from observations (observation nudging) or from gridded data sets (grid nudging) like global reanalysis fields. The reanalysis fields contain numerous observations that are constrained by models to give physically consistent fields of the atmospheric state variables.
Because our research is focused on long term reconstructions of the distribution of persistent pollutants, continuous grid nudging is the appropriate nudging type for the calculation of the meteorological fields. In this paper we investigate the influence of different nudging options in terms of the nudging of different state variables in different height ranges of the atmosphere on the results of a long term CMAQ model run for aerosols and persistent pollutants that are bound to them (here benzo(a)pyrene). Additionally, the influence of a land surface model and of lower vertical resolutions of the model is tested. The computing time for the MM5 simulations could be strongly reduced if simulations with lower vertical resolution would lead to sufficiently accurate results.

2 Model

CMAQ has been developed under the leadership of the Atmospheric Modeling Division of the Environmental Protection Agency (EPA) National Exposure Research Laboratory in Research Triangle Park, North Carolina, USA. The modeling system and its source codes are freely available for use by air quality regulators, policy makers, industry, and scientists to address multiscale, multi-pollutant air quality concerns. It includes a chemistry transport model that currently allows for the simulation of concentrations and deposition of the major air pollutants and particulate matter. Because of its generalized coordinate system and its advanced nesting features CMAQ can be used to study the behavior of air pollutants from local to regional scales. A detailed description of the model system is given by Byun and

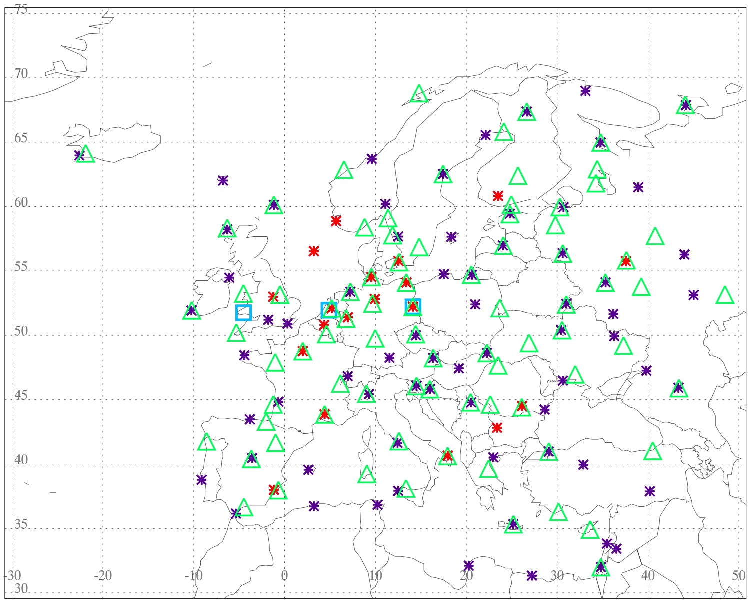

Fig. 1 Map of the selected measurement stations. Red stars: Selected radiosonde stations for the test of the different schemes. All stars: All radiosonde stations for the test of the regional and seasonal dependence of the results. Blue squares: Wind profiler stations. Green triangles: Precipitation measurement stations.

Ching [4] and more recently by Byun and Schere [5].
The model has been expanded at GKSS to study the trans-boundary transport of polycyclic aromatic hydrocarbons (PAHs; i.e. benzo(a)pyrene) and their deposition within coastal regions of Europe [1]. The goal of our studies are multi-year runs of MM5-CMAQ for the assessment of past trends in PAHs concentrations and deposition. The model is set up on a 54 x 54 km^2 grid for Europe and on a nested smaller domain with a 18 x 18 km^2 grid for the North Sea region.
MM5 is widely used and tested in the scientific community (see e.g. [7, 11]), it serves as preprocessor to derive the meteorological fields for several chemistry transport models, e.g. CHIMERE [2, 20], EURAD [14, 18], and CMAQ. In this study MM5 is operated with the more sophisticated parameterizations for cloud micro physics (Reisner2) [19], the planetary boundary layer (MRF) [13], and the subscale cumulus convection (Kain Fritsch 2) [15]. The Noah land surface module (LSM) [6] is used and the model is driven by ERA40 reanalysis data (1 x 1 degree, 6 hourly atmospheric fields, surface and soil data). FDDA is used by grid nudging the ERA40 data in different configurations.

3 Measurement data

The modeling of persistent pollutants, that are only present in very low concentrations in the atmosphere, aims at long times series over several years. Therefore the meteorological fields were tested for systematic deviations from long term observations. Because the long range transport of atmospheric pollutants is closely linked to its vertical transport, it is important to compare the model results to data that contains also vertical information and not only to ground data.

Radiosoundings that are routinely carried out by the European Weather Services and that can be publicly accessed are well suited for this purpose. The data comprises regular observations (usually twice a day) of temperature, wind, and humidity up to the tropopause. One disadvantage is of course that the data is already assimilated in the driving reanalysis fields. However there is no real alternative to the data if a homogeneous data set covering whole Europe is needed. All radiosoundings used in this study were extracted from the Integrated Global Radiosonde Archive (IGRA) data set [9], that contains data from more than 1000 stations world wide in a common format.

Nevertheless, we used also wind profiler data to check wind speed and wind direction at three stations in Central Europe. This data stems from the CWINDE network of windprofilers [3] that has grown to more than 25 stations in Europe in 2007. The data typically consists of hourly or half-hourly wind speed and direction in 20 to 30 layers between 200 m and 5000 m agl. Some instruments are constructed to cover the middle and upper troposphere with a height range between approx. 2000 m and 10000 m. The higher temporal resolution is a clear advantage against the radiosonde data. However, the data often includes some longer gaps that are mostly related to technical problems or maintenance of the instruments. In 2000, the year of interest for the CMAQ simulations, the network was in its development phase and included about ten stations.

Precipitation data from the European Climate Assessment (ECA) [16] was used for the comparison of the modelled amount of rainfall and the number of days with rainfall to the observations.

Fig. 1 shows the locations of 88 stations in Europe that were selected for our tests. A subset of 19 stations was used for the determination of the best nudging options while the whole set was used to determine regional or temporal inhomogeneities of the model results. The location of the three wind profiler sites and the 82 precipitation sites can also be seen in Fig. 1.

4 Results

The testing of the influence of the nudging schemes on the CMAQ results for particle bound pollutants and particularly benzo(a)pyrene was done in three steps. First, MM5 was run for one month, April 2000, in nine different configurations and the results were compared to radiosonde observations at 19 selected stations with a focus the North Sea region. This is the region of interest for the future use of the model at the Institute for Coastal Research at GKSS. The episode was chosen sufficiently short to allow a number of different model runs in a reasonable time, but long enough to include different weather situations. Second, MM5 was run for the entire year 2000 with the optimal configuration that was determined in the first step. For this year a detailed emission inventory for anthropogenic pollutants [10] and for B(a)P [8] is available. The results were then compared to measurements from 88 radiosonde stations to investigate possible problems in particular regions in Europe or in particular seasons. Last, the effects of the optimal nudging procedure on the aerosol and the benzo(a)pyrene concentration and deposition patterns in Europe were investigated and compared to the case when no nudging was applied.

4.1 Nudging Schemes

To test the different nudging schemes, several runs with different options were performed for April 2000 and the results were compared to the measurements at 19 selected stations. This month was chosen because it represents more or less average conditions in terms of temperature and humidity. The weather situation in Europe was mixed with low pressure systems approaching the continent quite frequently from the west in the first twenty days of the month. In the last third central Europe was influenced by the advection of warm air masses from the south. Nine different model setups were chosen for the initial tests:

1. NN: No nudging
2. PR: Periodic restart of the model every 96 hours, no nudging
3. UV: Wind (U,V) nudging
4. UVT: U,V and temperature (T) nudging
5. FN: U,V,T and water vapour mixing ratio (MR) nudging (full nudging)
6. noLSM: U,V,T,MR nudging, no LSM
7. vSST: U,V,T,MR nudging, daily varying sea surface temperature (SST)
8. 9L: U,V,T,MR nudging, only 9 vertical layers
9. 12L: U,V,T,MR nudging, only 12 vertical layers

All runs except case PR spanned a time series of 34 days (from 28 March 2000 to 30 April 2000) without restart. The first 4 days were considered as spin up and the results of 1 April to 30 April were then used for the comparisons. Only for the periodic restart, the model was run for 11 periods of 4 days each, using only the last 3 days for the comparisons. No further variations of the default values of the nudging coefficients in MM5 were applied. The coefficients determine how strong the influence of the nudged data (here the reanalysis data) on the model results will be. They were $2.5 \cdot 10^{-4}s^{-1}$ for wind and temperature and $1.0 \cdot 10^{-5}s^{-1}$ for the water vapor mixing ratio. Both temperature and water vapour mixing ratio were not assimilated in the PBL. The sea surface temperature (SST) is usually initialized at the beginning of a MM5 run and then kept constant over the whole run. In setup vSST, the SST was daily adapted to the SST given in the reanalysis data.
The model results were compared to the vertical profiles of wind, temperature and humidity derived from the radiosondes. For this purpose the radiosonde data was linearly interpolated to the model levels. The modeled profile was determined by considering the data from the four grid cells closest to the location of the radiosonde station. The four profiles were weighted with an inverse distance method and then averaged separately for each level. Drifts of the radiosonde during the ascent were not taken into account. At each station, the mean difference and the root mean square (rms-)error were calculated for each profile and these were then averaged for the whole month. Because the radiosonde data is typically available once or twice a day, this leads to 30 - 60 profiles per month. If less than 10 values per month were available in a certain level at a certain station, this data was not considered for the monthly mean. The results for temperature and relative humidity are displayed in Figures 2 and 3.
It can clearly be seen that nudging of the temperature results in a significantly lower rms-error and in a very small mean difference between model and observations (Fig. 2). If no nudging is applied or only the wind components are nudged, the temperatures are underestimated by the model. The effect is less severe when the model is restarted every 4 days, but the results are still significantly worse than for the nudging cases. The number of vertical layers that was used in MM5 has also an important effect. The simulations with 9 and 12 vertical layers showed the largest rms-errors and the temperatures were on average 1 K

too high. The use of the land surface model and the daily varying SST do not significantly influence the results, which can be interpreted in a way that the nudging of the temperature already determines the performance of the model.

The picture is slightly different for relative humidity (Fig. 3). Again, highest deviations from the observations were detected for the cases without nudging. All other results did not deviate considerably from each other and even if only the wind components U and V were nudged, the rms-error and the mean deviation were much lower than for the cases without nudging. The use of a land surface model slightly improved the results. The lower number of vertical layers in the setups 9L and 12L did not lead to higher mean deviations or rms-errors. On the contrary, these simulations showed the lowest rms-errors. However, at all stations the modeled values of the relative humidity were about 5 to 10 % higher than the observations.

For wind direction and wind speed, the two runs without nudging also showed much higher rms-errors than all other runs (Figures not shown). Though, the mean deviations of wind speed and wind direction were similar for all runs.

Because the main sink for aerosol particles and therefore also for all pollutants connected with them is wet deposition, also the precipitation results were compared with measurements at 82 stations in Europe. Table 1 displays the results for April 2000. Average monthly rainfall, average rain days (with more than 1 mm rain) and the number of stations where the mean deviation of the daily rainfall was below 1 mm are reported. The cases without nudging (NN and PR) showed the worst agreement in the number of average rain days (on average 2.4 and 3.6 more rain days) and the most stations with large differences from the measurements (25 and 29 out of 82 stations). Despite these facts the average monthly rainfall was captured quite well by the NN run but the PR case showed the highest rainfall (+ 17 % averaged over all stations). The runs with a reduced number of layers were the driest ones (lowest average rainfall (-20% and -26%) and lowest number of rain days). They showed the highest deviations from the average monthly rainfall. The runs with nudging showed the highest number of stations with less than 1 mm deviation of the daily mean precipitation and (except the runs with full nudging) also close agreement in the total monthly rainfall. Among the runs with nudging, the FN run came closest to the average rain days but it showed about 11 % less precipitation than the measurements.

In summary, it can be said that nudging of all the variables U, V, T and MR leads to the lowest deviations from the observations. The results are only slightly influenced if MR is not nudged, on the contrary, the average rainfall was captured better without nudging the water vapour mixing ratio. The inclusion of a daily varying SST had almost no influence on the results. The humidity was slightly stronger overestimated when the land surface module was switched off. The choice of a much lower number of vertical levels for the model runs can have significantly adverse effects on the model results. In particular temperature and precipitation showed considerably worse agreement with the observations although a full nudging of U, V, T and MR was applied.

4.2 Seasonal and regional dependence

Additional to the dependence on the nudging scheme, which was tested only for April 2000, the model results might also depend on season and on location. Therefore, annual runs were performed with complete nudging of U, V, T and MR, the use of the Noah LSM and monthly varying SST. Each month was modeled separately with a spin up time of 4 days. Separate sensitivity tests have shown before that this time is sufficient to achieve results that are almost independent from the initial conditions. The results are displayed in color code in Fig.

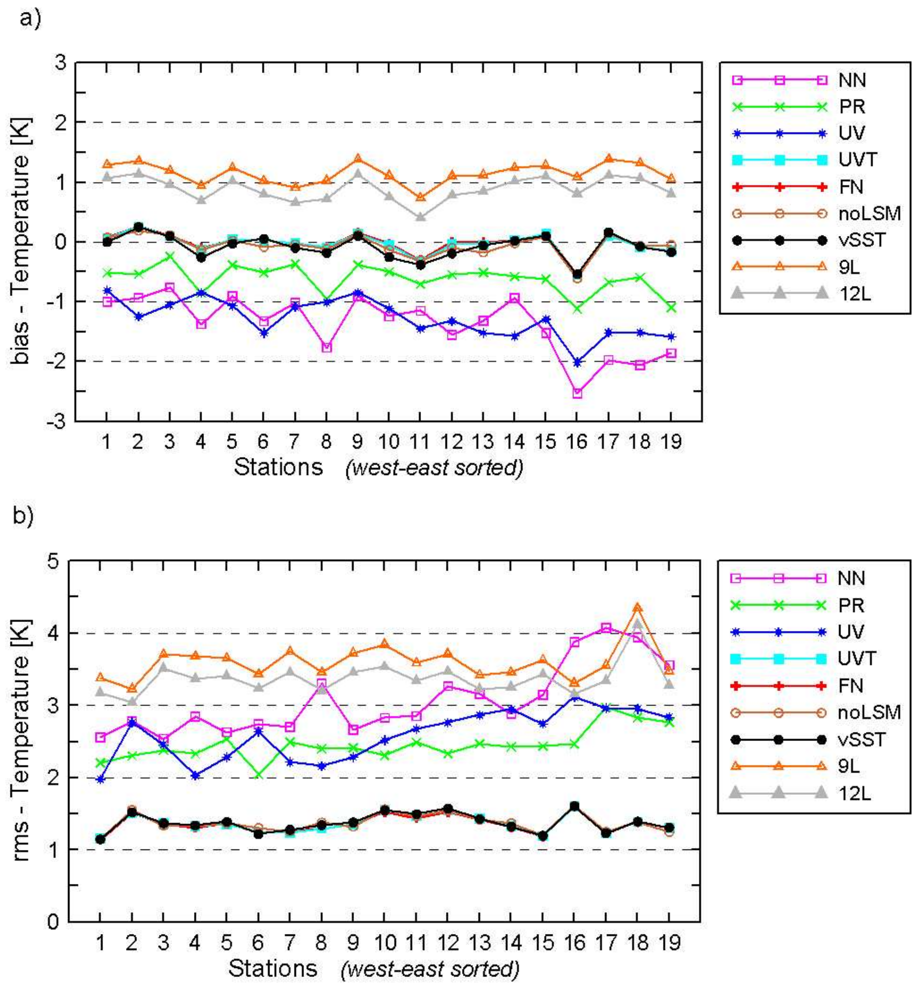

Fig. 2 Mean deviation (a) and mean rms error (b) of the modeled temperature compared to radiosoundings at 19 selected stations in Europe in April 2000.

Table 1 Comparison of simulated and measured rainfall at 82 stations from the ECA data set in April 2000.

case	average monthly rainfall mm	average rain days #	stat. w. m. daily diff. $<$ 1.0 mm #
measured	**58.9**	**9.9**	**82**
NN, no nudg.	59.9	12.3	53
PR, periodic	69.0	13.5	57
UV, U,V nudg.	60.6	11.9	64
UVT, U,V,T nudg.	57.5	11.4	65
FN, U,V,T, MR nudg.,	52.4	10.7	64
noLSM, f. nudg., no LSM	57.4	11.3	64
vSST, f. nudg., var. SST	53.1	10.8	66
9L, f. nudg., 9 lay	43.5	8.8	61
12L, f. nudg., 12 lay.	47.4	9.6	63

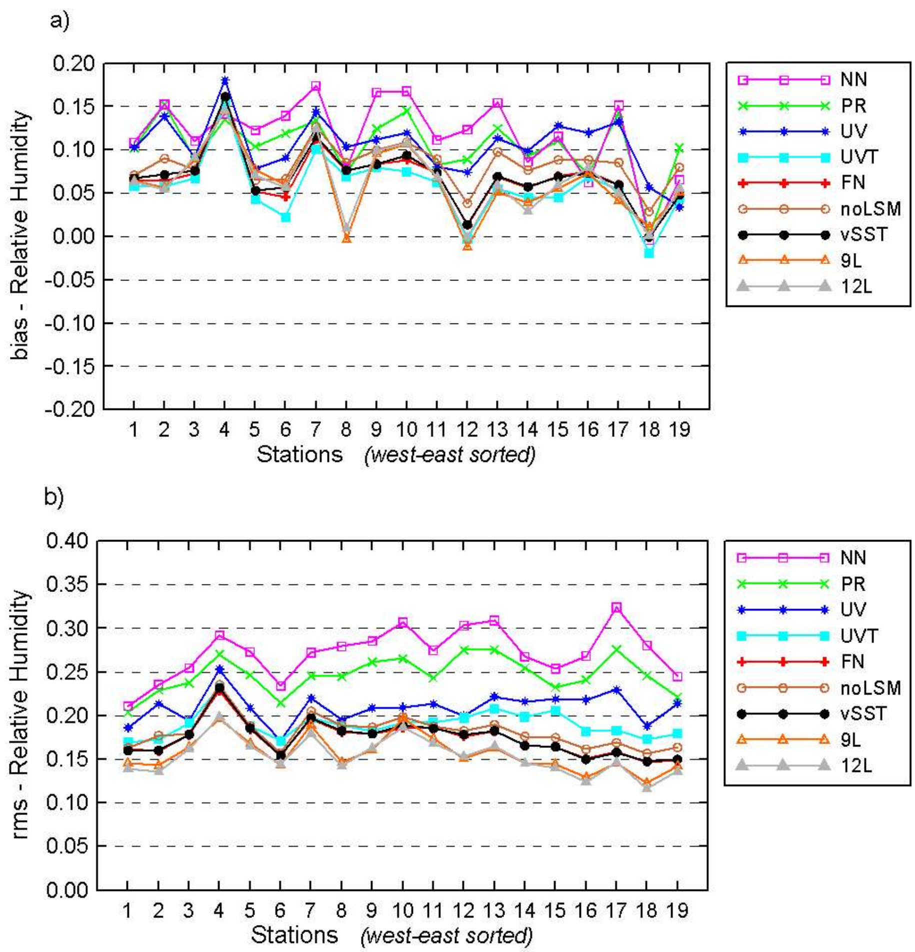

Fig. 3 Same as Fig. 2 but for relative humidity.

4 to 7. The x-axis shows the temporal variation and the y-axis shows the stations which were sorted from west (No.1) to east (No.88). Here, only the bias in temperature, rel. humidity, U-wind component and V-wind component are given. Black colors denote not enough data. Some scatter in the results for the temperature can be seen in Fig. 4, but no systematic effects were present. Some stations (in particular Reykjavik, the most westerly station in the model domain) show large discrepancies between model and observations but the absolute bias is usually below 0.5 K.

Some seasonal and regional effects were found for the relative humidity. While in winter and spring, RH is mostly overestimated (as it has already been seen for the April results), some underestimations are also observed in summer and fall at the more easterly stations. Nevertheless, the results give a quite uniform picture.

The results for the wind components show quite some scatter among the stations and the different months but no large effects in particular seasons or regions. While the U-wind component is on average slightly overestimated, the V-wind component is slightly underes-

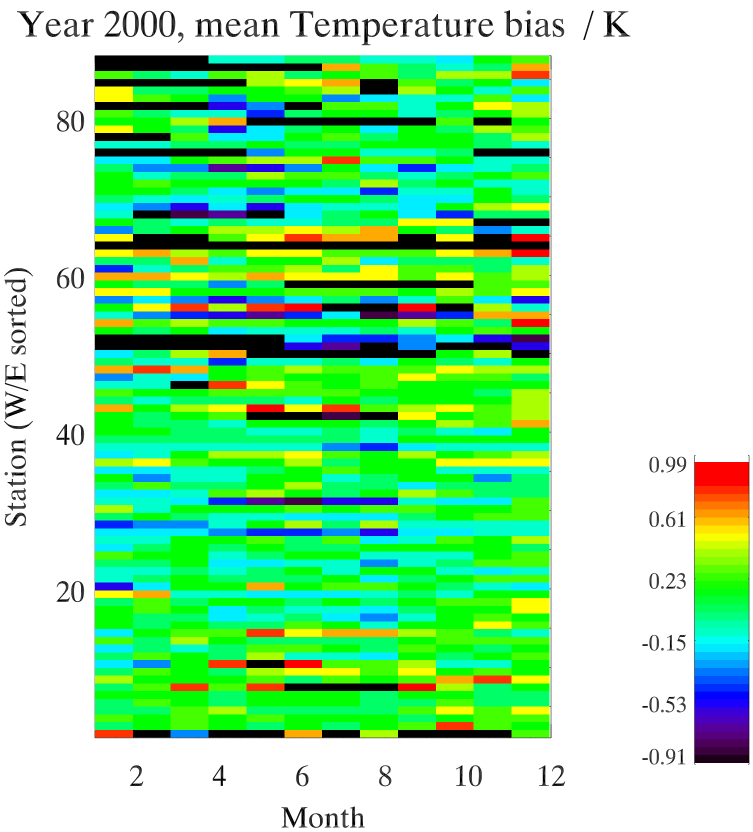

Fig. 4 Monthly comparison of the mean deviation of temperature (in K) from radiosonde profiles at 88 European stations and MM5 model results calculated with complete nudging.

timated.

Up to now the model results have been compared to data from radiosonde stations. Their advantage is that they are taken on a regular basis with proven data quality and they yield vertical profiles of the main meteorological parameters. However this data was already used to construct the reanalysis fields that served as initial and boundary conditions and as data for the nudging procedure. Therefore it is worth to compare the data with an additional independent data set. Because the wind data is of utmost importance for a transport model this data was also compared to hourly wind profiler data at three European stations of the CWINDE wind profiler network [3]: Pendine/United Kingdom, Cabauw/The Netherlands and Lindenberg/Germany (Fig. 8 to 10). In this case, the annual time series at all model levels where profiler data was available were investigated. At Pendine, a slightly negative V-wind bias was observed, the U-wind bias was slightly positive over the whole year. The absolute deviations increase with wind speed and therefore with altitude. The temporal correlation of the time series is higher than 0.9 in all levels, in altitudes above 1 km higher than 0.95. The bias is slightly lower at Cabauw, here V is also underestimated while U is

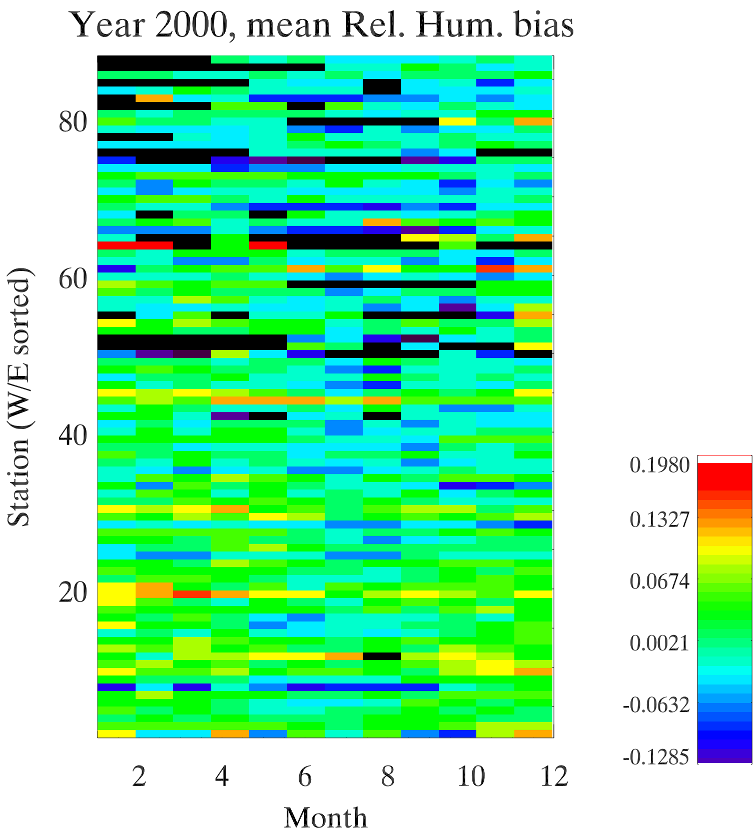

Fig. 5 Same as Fig.4 but for the relative humidity.

overestimated. The temporal correlations are not as high as at Pendine, they vary between 0.8 and 0.9. Closest agreement was achieved at Lindenberg, where the bias is almost zero and the correlations are around 0.95.
Although wind profiler data for the year 2000 was available in sufficient completeness only at a few stations, the results of the comparisons between model simulations and radiosondes and the close agreement could be confirmed by these local tests.

4.3 Influence on CMAQ model results

The influence of the different model setups on the secondary inorganic aerosol (SIA) and the B(a)P concentrations was investigated for two of the nine different cases described in section 4.1. The reference run was performed with full nudging (FN) and it was compared to the case when the model was periodically restarted (PR) but none of the variables was nudged. A comparison of the B(a)P model results to atmospheric measurements is difficult because the data is sparse and if it is available only monthly averages are provided. This

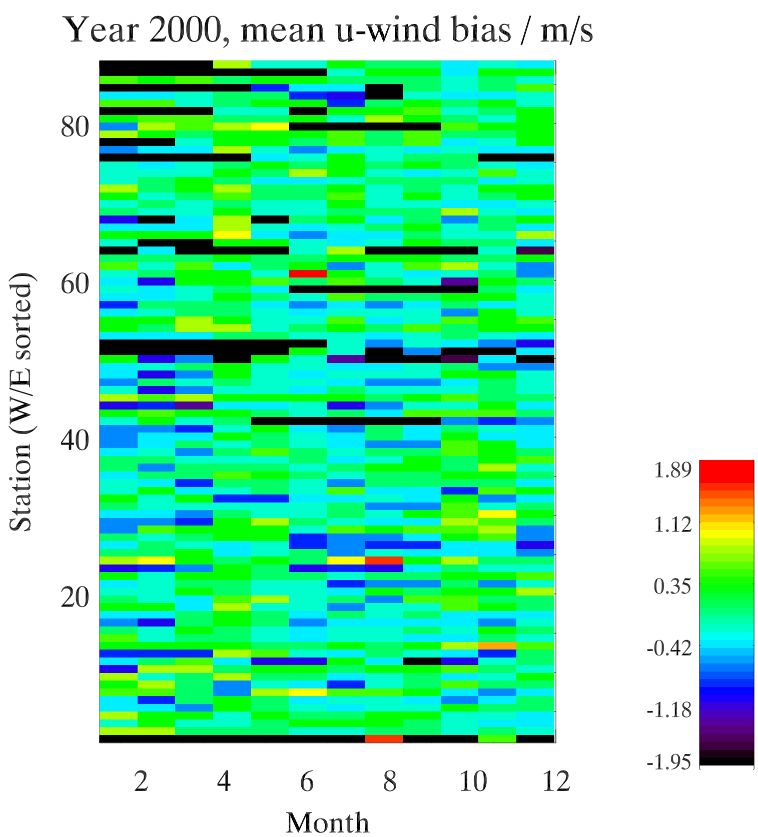

Fig. 6 Same as Fig.2 but for the u-wind (in m/s).

is caused by the low B(a)P concentrations in ambient air, that are in the order of less than 1 ng/m^3, and by the fact that no automated procedures to measure B(a)P on filter samples are proven. B(a)P is mostly bound to aerosol particles. Therefore, the ability of the model to simulate atmospheric transport of secondary inorganic aerosol can be used as a measure to test the differences in the model results if the different meteorological fields are applied. The aerosol components sulfate (SO_4), nitrate (NO_3) and ammonium (NH_4) are measured on a daily basis at several measurement stations of the EMEP network [22] and they are freely available (`www.emep.int`). A comparison of the monthly mean concentration of particle bound sulfate at 20 EMEP stations with the modelled values for the two runs is displayed in Fig. 11. Generally, the sulfate concentrations are underestimated, particularly in Northern Germany and Denmark (stations DE07 - DK08). This is possibly caused by the fact that ship emissions, that contribute significantly to the sulfur emissions in North Sea coastal regions, were not included in the emissions data base. It can clearly be seen that the model run with complete nudging exhibits higher sulfate concentrations and on average better agreement with the measurements than the model run where the meteorological model was only restarted every 96 hours. Averaged over all stations, the mean sulfate concentration

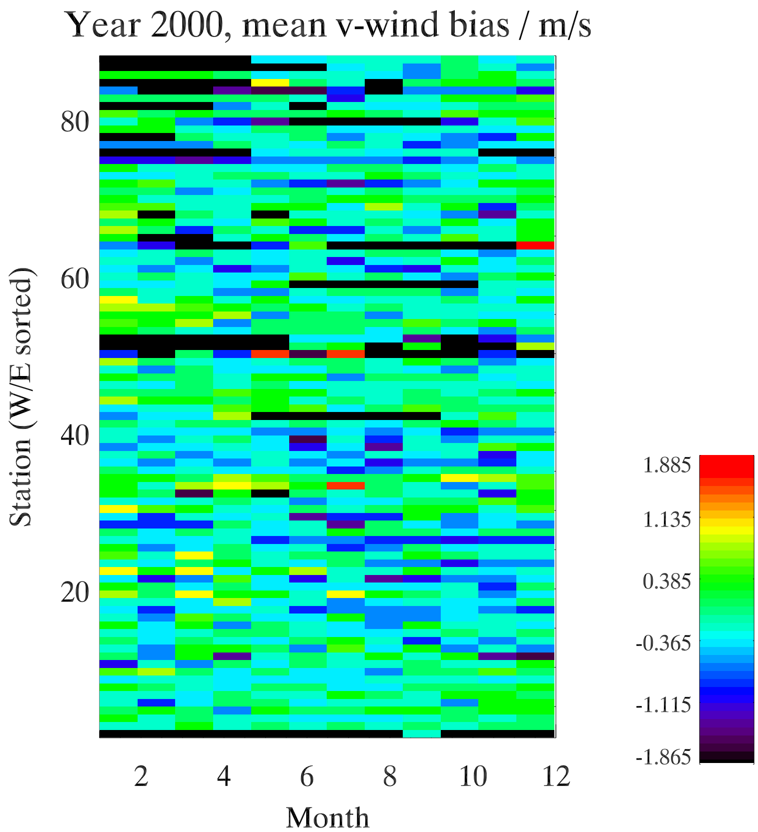

Fig. 7 Same as Fig.2 but for the v-wind (in m/s).

in April 2000 was 2.94 $\mu g/m^3$. The PR run showed 1.91 $\mu g/m^3$ while the FN run showed 2.23 $\mu g/m^3$. The results of the FN run were closer to the measurements at 15 out of 20 stations. The index of agreement (IOA) increased on average from 0.54 to 0.61. It was higher at 13 out of 20 stations in the FN run. The same comparisons were also done for nitrate and ammonium, however for these species only data from 8 stations was available. On average ammonium was captured quite well in both configurations. The measurements gave a mean of 1.35 $\mu g/m^3$, the PR run showed 1.21 $\mu g/m^3$ while the FN run resulted in 1.42 $\mu g/m^3$. The IOA increased on average from 0.59 to 0.62 and it was higher at 5 out of the 8 stations. In the case of nitrate the comparison led to a clearer picture. Here the average measured value was 2.99 $\mu g/m^3$, the FN run gave 1.90 $\mu g/m^3$ while the PR run showed only 1.52 $\mu g/m^3$. The average IOA increased from on average 0.48 to 0.56 in the FN run, but again it was higher at only 5 of the 8 stations compared to the PR run. A detailed investigation of the CMAQ model results for the simulation of SIA for the years 2000 and 2001 can be found in Matthias (2008) [17].

The consequences of the different nudging procedures on the B(a)P simulation results are shown in Fig. 12 for the mean concentration at ground level in April 2000 and in Figure

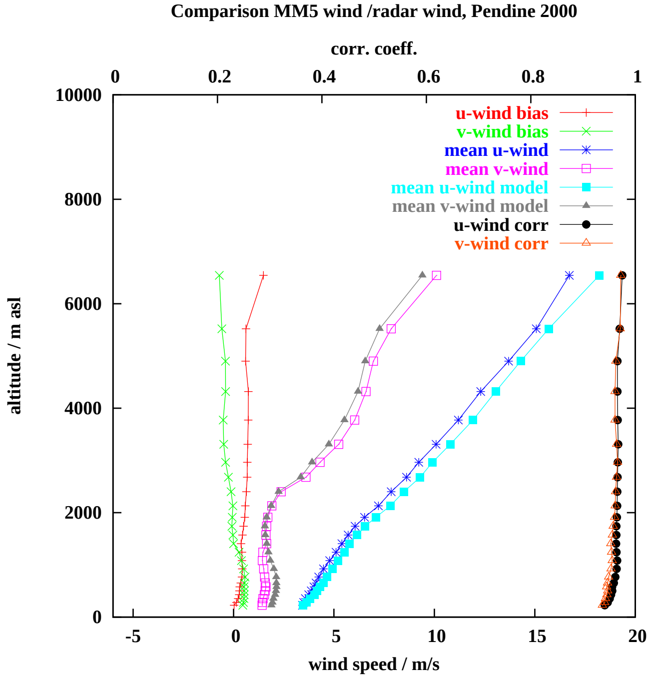

Fig. 8 Comparison of U- und V-wind components calculated with MM5 for 2000 with hourly wind profi ler data at Pendine/UK.

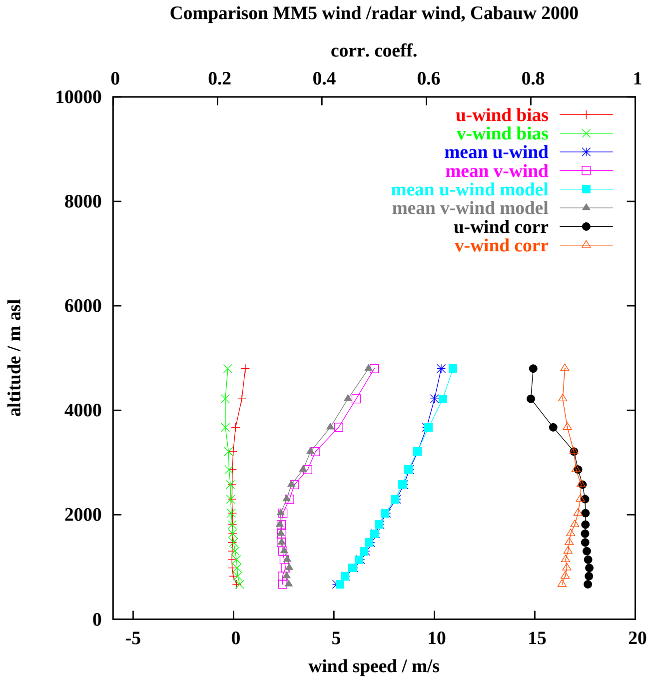

Fig. 9 Same as Fig.8, but at Cabauw, The Netherlands.

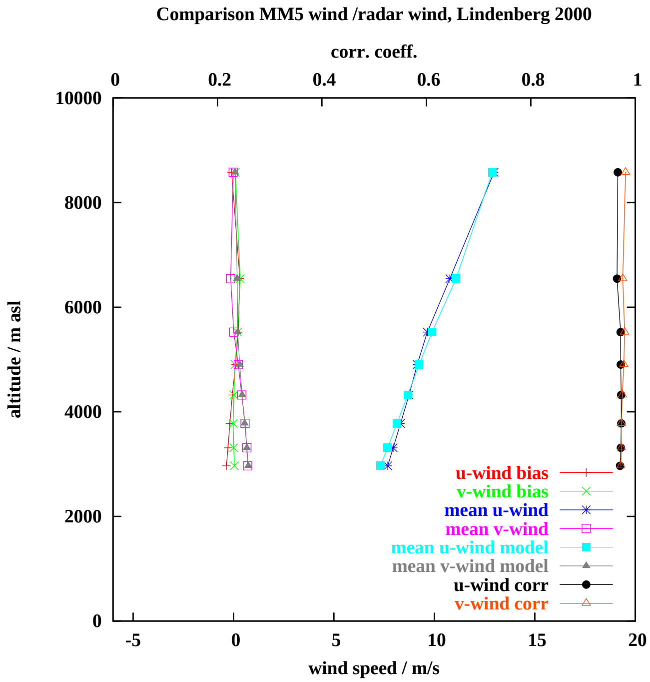

Fig. 10 Same as Fig.8, but at Lindenberg, Germany.

13 for the mean wet deposition. In the PR run, lower concentrations were modeled in some regions (like the Po basin in north Italy, in central Europe and around Moscow). In these regions, higher wet depositions occur which leads to the conclusion that changes in the hydrological cycle might cause the differences in concentrations. This is also reflected in the remarkably high rainfall rate of the PR run. It is additionally interesting to note that in the FN run, high B(a)P depositions over the North Sea are simulated while this feature is much less pronounced in the PR run. The same holds for high depositions in central France and southern Scandinavia. On the other hand the high B(a)P deposition in south Finland in the PR run is not visible in the FN run. Comparisons of the results to B(a)P measurements cannot be used here to estimate the model performance because they are very sparse and are typically only given as monthly averages.

5 Conclusions

The influence of different nudging options and the vertical resolution in the mesoscale meteorological model MM5 on the SIA and the B(a)P concentrations and depositions in April 2000 was tested. Out of the nine cases that were investiagted with respect to the meteorological variables T, RH, U and V, the meteorological fields calculated with complete nudging of these variables showed the lowest deviations from regular radiosonde observations. Selected wind profiler time series were also compared to the modeled wind field. The close agreement between the model results and the measurements could be confirmed by this independent data set.

A lower number of vertical layers in MM5 lead to a considerably worse agreement of the

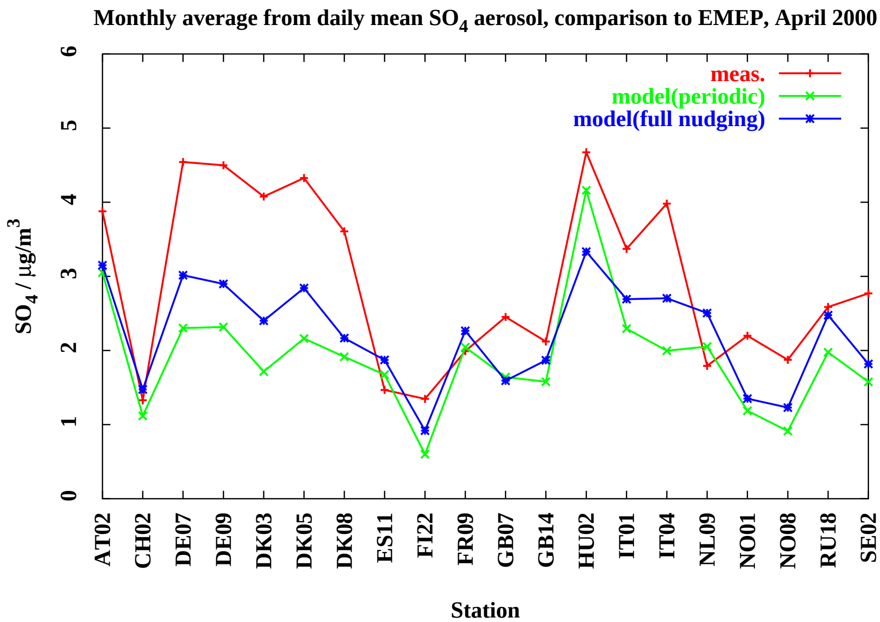

Fig. 11 Monthly mean particle bound sulfate concentration at 20 EMEP stations in April 2000.

model results, particularly temperature and rainfall, with the observations. The temperature was on average too high while the rainfall was too low in the reference time period.
The CMAQ results with full nudging showed considerably better agreement with measurements of particle bound sulfate, nitrate and ammonium compared to the case when no nuding was applied but the model was resarted every 96 hours. Significant differences in the B(a)P distribution over Europe, particularly in regions with high B(a)P concentrations were observed. Because in regions with lower B(a)P concentrations, enhanced wet deposition was modeled, it is likely that the hydrological cycle is significantly different in the two simulations. This is stressed by the fact, that comparisons with precipitation measurements at 82 European stations revealed sigificantly higher rainfall in the PR case.
Finally, it can be stated that for hindcast studies a nudging of the variables T, U and V is highly recommended while a nudging of the mixing ratio does not lead to clearly better results. However if a low number of vertical layers is chosen in the meteorological model, the model results might differ considerably from the observations even if a full nudging was applied.

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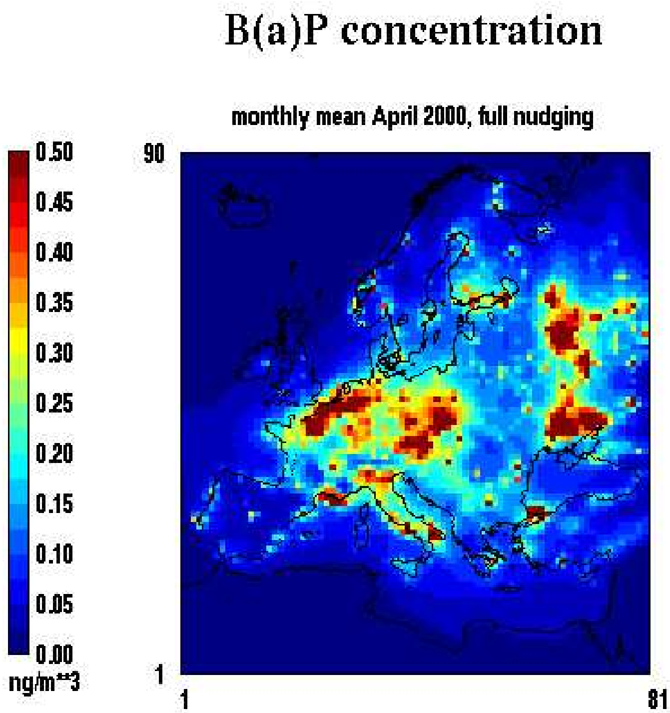

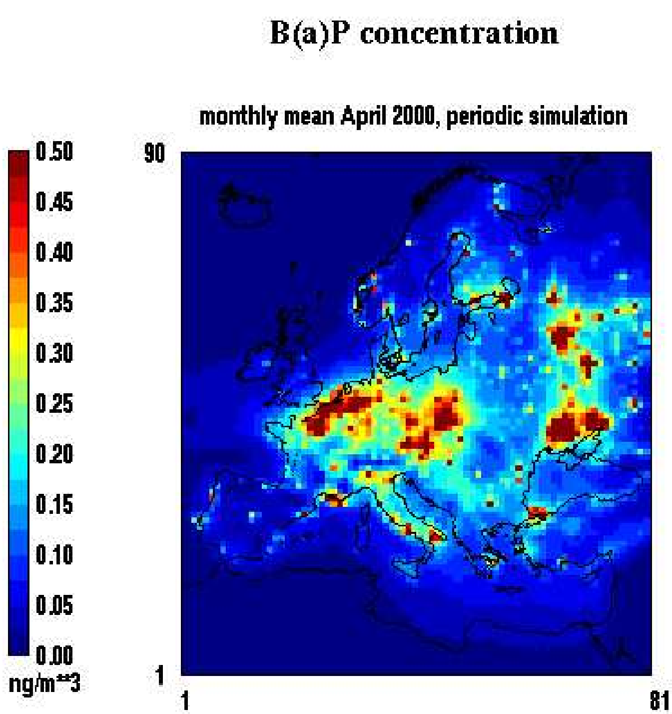

Fig. 12 Modeled B(a)P concentrations in Europe in April 2000 calculated with different meteorological input fields. Top: with full nudging of T, MR, U and V. Bottom: without nudging but with a periodic restart of the model every 4 days.

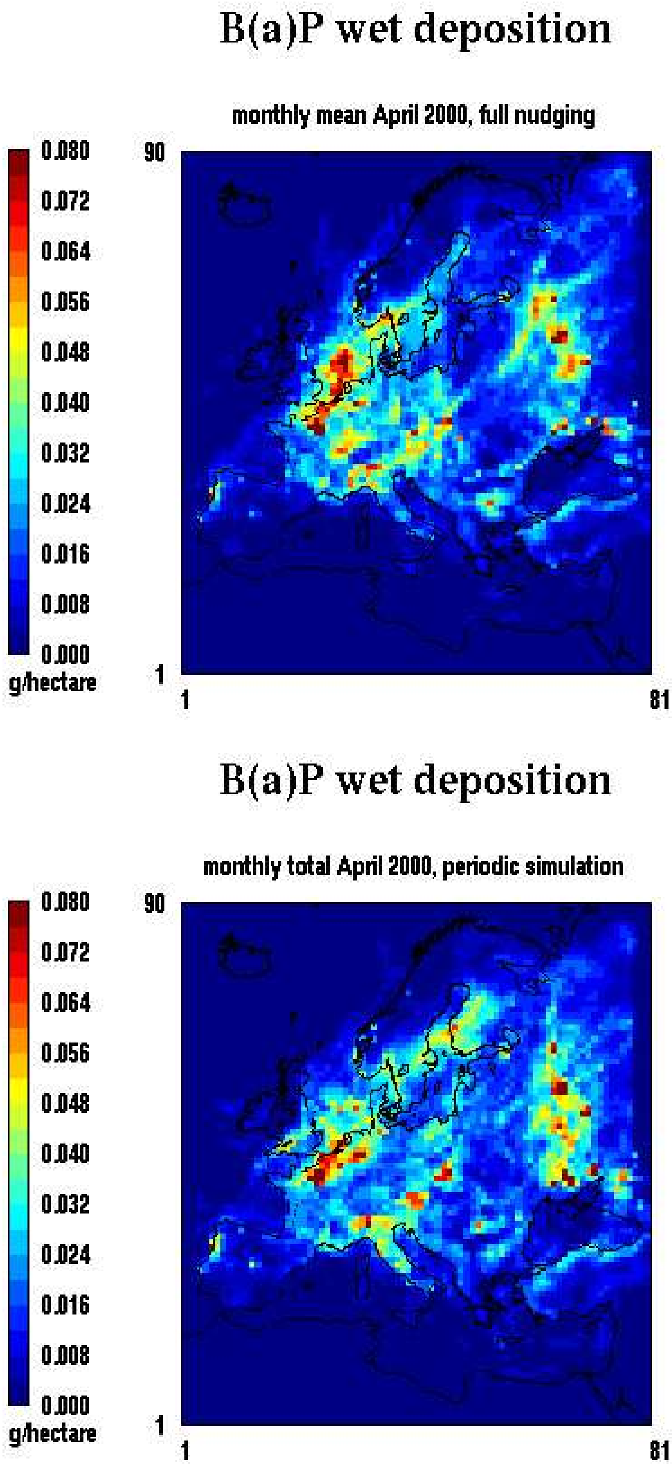

Fig. 13 Same as Fig.12 but for wet deposition.

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