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Discussions

Impact of the vertical emission profiles on ground-level gas-phase pollution simulated from the EMEP emissions over Europe

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Abstract

Five one-year air quality simulations over a domain covering Europe have been performed using the CHIMERE chemistry transport model and the EMEP emission dataset for Europe. These five simulations differ only by the representation of the effective emission heights for anthropogenic emissions: one has been run using the EMEP standard recommandations, three others with vertical injection profiles derived from the EMEP recommandations but multiplying the injection height by respectively 0.75, 0.50 and 0.25, while the last one uses vertical profiles derived from the recent literature. It is shown that using injection heights lower than the EMEP recommandations leads to significantly improved simulation of SO_2, NO_2 and O_3 concentrations when compared to the Airbase station measurements.

1 Introduction

Air quality modelling has emerged in the recent decades as an important element in understanding and forecasting chemistry in the troposphere, particularly over highly urbanized and industrialized regions as it is the case in Europe. While this was first performed at the urban scale, it has been shown that a proper representation of long-range transport of ozone and its precursors need to be considered in order to have realistic modelling of air quality, even at urban scale. Therefore, eulerian chemistry-transport models such as CHIMERE, CMAQ or CAMx, among others have been developed since the 1990s and now typically include anthropogenic emissions, biogenic emissions and advanced chemistry such as MELCHIOR or SAPRC.

The three major ingredients of air quality modelling are a meteorological simulation adequately representing the state of the atmosphere in the considered region, anthropogenic and biogenic emission data for each model grid cell, and a chemistry-transport model. The model results can then be validated using ground measurements (station data, LIDAR, dropsondes) or satellite measurements.

Generally, anthropogenic emission data are obtained from a top-down strategy, i.e. downscaling national emission totals horizontally, vertically and in time to provide these emissions to chemistry transport models. The horizontal disaggregation is generally realized through the use of proxies such as landuse, population density or transportation network, the temporal disaggregation follows seasonal and sub-diurnal disaggregation factors depending on the countries. The emission data used for this study are taken from the EMEP gridded emission dataset at 0.5° horizontal resolution (Vestreng, 2003; Vestreng et al., 2009).

Vertical disaggregation, i.e. estimation of the effective emission heights for anthropogenic emissions, is either realized using plume-rise models such as SMOKE (Bieser et al., 2011) or tabulated factors depending on the SNAP sector considered. In the latter case, EMEP-provided disaggregation factors as provided in, e.g., (Bieser et al., 2011) are the most commonly used. As noted in Bieser et al., 2011, these profiles are based on plume rise calculations for the city of Zagreb, and may not be representative of other European regions. Furthermore, other studies (De Meij et al., 2006; Pregger and Friedrich, 2009; Bieser et al., 2011) have questioned these results using other methodologies, obtaining effective emission heights lower than the EMEP recommendations. Pregger and Friedrich, 2009 have used data from 12699 industrial stacks in 10 German Federal State, from the IER inventory, which were aggregated in 34 categories, for which relevant parameters such as weighted and unweighted average, median and standard deviation are provided for 4 main parameters: stack height, flue gas temperature, flue gas velocity and flue gas flow rate. For each of these source types, they also calculated effective emission heights assuming a standard atmosphere, finding effective emission heights significantly lower than the EMEP recommendations. Following this work and using the database provided for stack characteristics (assuming that these characteristics are relevant for all Europe), Bieser et al., 2011 used the SMOKE-EU model to calculate 44 976 vertical emission profiles for Europe depending on SNAP sector, country, climate zone, season, day and night, and pollutant type. These 44 976 profiles have then been reduced to 73 using cluster analysis which they use as input for the CMAQ CTM, showing that the use of these 73 profiles yielded significantly stronger SO_2 and sulfate concentrations near the ground than when using the EMEP profiles. Finally, these authors provide new emission profiles following the classical EMEP layering (their Table 3). This direct comparison to the EMEP recommendations confirms the conclusions of De Meij et al., 2006 and Pregger and Friedrich, 2009 regarding the fact that the latter are likely to overestimate the effective emission heights.

At global scale, Pozzer et al., 2009 have performed a sensitivity study on vertical distribution of anthropogenic emissions using the atmospheric chemistry general circulation model EMAC (ECHAM/Messy atmospheric chemistry, Jöckel et al., 2006), performing two simulations at T42 spectral resolution, corresponding to approximately $2.8^\circ \times 2.8^\circ$. The control simulation is performed by affecting the anthropogenic emissions to 6 layers between 45 m and 800 m above ground level, using fixed vertical profiles per emission class and species, and the test simulation is performed affecting all anthropogenic emissions (except aircraft emissions) to the lowest model layers. The authors show that the effect of this vertical redistribution is strong particularly for NO_x, CO, NMVOCS and O_3. It is worth noting that the above-mentioned study did not evaluate the impact of the vertical distribution on SO_2 emissions, even though SO_2 can be considered the most sensitive species to vertical emission profiles (Bieser et al., 2011). It is also worth noting that, contrary to CHIMERE, the ECAM model includes the feedback of chemistry on meteorology through radiative processes, so that the two chemical simulations are not performed with strictly the same meteorology. These effects yield differences up to 15 % in specific humidity, however, due to nudging to the ECMWF operational analysis data, the authors indicate that this impact is very weak at least for long-term averaged values.

Despite this renewed interest during the recent years in the estimation of effective emission heights however, to the authors' knowledge, no study has systematically investigated the impact of updating the EMEP emissions heights towards other vertical profiles in a study validated through comparison with real-world data. The purpose of the present paper is to examine several strategies for revising the EMEP vertical disaggregation, either performing manual adjustments from the EMEP profiles or vertical profiles adapted from the Bieser et al., 2011 study, and evaluate the impact of these updated vertical profiles on CHIMERE performance relative to Airbase measurements over Europe.

2 Data and methods

2.1 Models and configuration

The results presented in this version have been obtained with the CHIMERE 2011 version (Menut et al., 2013). CHIMERE is an off-line chemistry-transport model (CTM), which models atmospheric chemistry and transport, forced by anthropogenic emissions, biogenic emissions, a meteorological simulation and boundary conditions. First developed in 1997 as a box model covering the Paris area with only gas-phase chemistry (Honoré and Vautard, 2000; Menut et al., 2000; Vautard et al., 2001), it is now a cartesian-mesh grid model including gas-phase, solid-phase and aqueous chemistry (Bessagnet et al., 2004), biogenic emissions modelling depending on meteorology with the MEGAN model (Guenther et al., 2006), dust emissions and resuspension (Menut et al., 2005; Vautard et al., 2005). CHIMERE has been evaluated against measurements and other CTMs both at urban scale (Vautard et al., 2007; Van Loon et al., 2007; Schaap et al., 2007) and at continental scale (Solazzo et al., 2012).

The simulation has been performed for a domain covering Europe at 0.5° resolution (Fig. 1), with 79 × 47 horizontal grid cells. This horizontal resolution permits a representation of the large-scale circulation and main patterns of atmospheric chemistry over Europe, but does not allow to represent small-scale effects such as the local effect of a road, a urban area or a factory. The vertical discretization is 8 vertical levels of increasing thickness away from the ground defined in hybrid sigma-p coordinates, with the first level at 0.997 sigma-level (about 25 m above the ground) and the top of the last level at 500 hPa. Even though many CTM tend to work with more vertical levels, for the case of

CHIMERE, the 8-level configuration is used successfully for operational prevision, and it has been shown recently that stepping up the number of vertical levels from 8 to 20 does not measurably improve the performance of CHIMERE at least in terms of values at ground level (Menut et al., 2012).

The simulation has been initialized with LMDZ-INCA climatological data for gas species and LMDZAERO for aerosols, also used as boundary conditions, the horizontal and vertical advection has been performed using the Van Leer I scheme (Van Leer, 1979). Atmospheric chemistry has been modelled using the MELCHIOR2 scheme (Derognat et al., 2003). Biogenic emissions have been generated using the MEGAN model. All the simulations presented in this study have been performed for the period from 20 February 2008 to 19 February 2009, in order to cover a complete annual cycle.

The meteorological simulation has been performed using WRF-ARW model (Michalakes et al., 2005; Skamarock and Klemp, 2008) version 3.2.1 on a 99 × 99 horizontal grid built with Lambert-conform projection with $45 \times 45\,\mathrm{km}^2$ horizontal resolution with a reference point at (49.115° N; 9.25° E) point, which is also the center of the domain. 27 vertical levels from 997 to 50 hPa have been represented. The WRF options used for this study are: Yonsei University Planetary Boundary Layer, WRF Single-Moment 6-class microphysics, RRTM (Rapid Radiative Transfer Model) longwave radiation, Dudhia simple downward integration shortwave radiation scheme, MM5 Monin-Obukhov similarity theory surface layer, Noah Land-surface model, Kain-Fritsch convection scheme. The WRF model used the boudary conditions provided by the global GFS analysis fields, and no nudging was applied. The output of the meteorological simulation has thereafter been interpolated by the CHIMERE model on its own horizontal lat-lon grid and hybrid sigma-p vertical levels using the prepmet and diagmet modules. The vertical component of the wind is recalculated after the projection from the divergence of the horizontal wind field in order to secure mass conservation.

2.2 Downscaling of the emissions

The CHIMERE horizontal grid for the simulations that have been performed correspond exactly to the grid cells for which the EMEP emissions at 0.5° × 0.5° are provided, so that no horizontal disaggregation is needed for this study, and the emission totals per species and per snap sectors are used directly as provided in the EMEP database.

The vertical downscaling of the EMEP emissions for CHIMERE, which is of interest for the present study, is performed in two steps. The first step is done during the preprocessing phase of the anthropogenic emissions in the emiSURF module, where the emissions are vertically distributed into the EMEP vertical layers as defined in the standard EMEP recommandations (Table 1). As the first EMEP layer is relatively thick (92 m), and includes more than one CHIMERE layer, to avoid unnecessary vertical dilution of surface emissions between several vertical layers, the EMEP profiles have been modified in the distributed version of CHIMERE, adding a supplementary 20m vertical level close to the ground. For SNAP sectors corresponding to surface emissions (SNAP 6, 7, 8, 10), 100 % of the emissions are affected to this 0–20 m layer, for other SNAP sectors, the 0–20 m receives a proportional share of the EMEP emissions from the 0–92 m. The temporal and spatial disaggregation of emissions is also performed at this stage as described in Menut et al., 2013. The second step of the vertical disaggregation of emissions occurs when the actual netcdf emission files covering the simulation period and interpolated on the model vertical grid are produced by the prepemis routine of CHIMERE. This routine affects the emissions from each EMEP layer to the corresponding CHIMERE layer, assuming uniform vertical repartition of the emissions within each EMEP layer.

Within this preprocessing, there are mainly two possible ways to interfere with the vertical distribution of the emissions. The first way is to adapt the EMEP recommendations by lowering or raising the EMEP layers, while the other way is to conserve the standard EMEP vertical layering but change the matrix attributing the emissions to vertical layers. In the present study, both these strategies have been explored: modifying the EMEP levels (by multiplication by a factor), and redistribute the emissions within the EMEP vertical layers following the Bieser et al., 2011 recommendations and adding an additionnal 0–20 m layer in a similar way as is done usually for EMEP emissions when used in CHIMERE. The disaggregation matrix per SNAP sector and EMEP levels obtained from the Bieser et al., 2011 recommendations and from standard EMEP procedure are recapitulated in Table 1. Even though Bieser et al., 2011 provides 73 different emission profiles depending on several parameters, they show that the main dependance is on SNAP sector, while other factors such as seasonal cycle, climate zone or day/night variations have a more modest impact according to this study. Therefore, as a first step, it has been chosen to use only the average profiles for each SNAP sector as provided in Table 3 of Bieser et al., 2011.

Following these lines, 5 different simulations have been performed for the considered period:

- CTL simulation: using the standard CHIMERE configuration, i.e. vertical disaggregation from Table 1 (top)
- h75 simulation: vertical disaggregation from CTL but lowering the altitude of the EMEP levels by multiplication by 0.75. The resulting EMEP levels are 159 m, 138 m, 243 m, 391.5 m, 585.75 m, 829.5 m
- h50 simulation: vertical disaggregation from CTL but lowering the altitude of the EMEP levels by multiplication by 0.5.
- h25 simulation: vertical disaggregation from CTL but lowering the altitude of the EMEP levels by multiplication by 0.25.
- Bie simulation: vertical disaggregation following Table 3 of Bieser et al., 2011 as presented in Table 1(bottom).

As a result of these modifications, the vertical effective emission profile for SO_2 substantially differs between the five performed simulations (Table 2). The CTL emissions displays the highest effective emission heights, with the h75, h50 and h25 displaying decreasing emission heights compared to the CTL simulation. The simulation based on the Bieser et al., 2011 profiles is relatively close to the h50 simulation, so that it is fair to say that, regarding SO_2 emissions, application of these recommendations lead to a downward reevaluation of the SO_2 effective emission heights of almost 50 % at the European level, which is considerable.

2.3 Observations and statistical methods

The observation data was obtained from the Airbase database. A total 2266 stations had NO_2 data for the covered period, 1688 had O_3 data and 1459 had SO_2 data. The 721 stations that have at least a 50 % coverage for the given period for the three above-mentioned trace gases have been selected for comparison with simulated values, which represent a total of 721 stations covering all the modelled area (Fig. 2).

Two criteria have been retained to compare the simulation outputs to observations. The mean bias of the model compared to the observations is calculated for each measurement station with sufficient data availability and then averaged over all stations or per station type and location. The skill score used in the present study is the same as the skill score *S* defined by equations presented in Mao et al., 2006, which are recalled here, for a given station with *N* time steps for comparison between model and observations:

$$\mathrm{BIAS} = \frac{1}{N}\sum_{k=1}^{N}\left(X^m - X^o\right), \quad (1)$$

$$\mathrm{ABSE} = \frac{1}{N}\sum_{k=1}^{N}|X^m - X^o|, \quad (2)$$

$$\mathrm{RMSE} = \left[\frac{1}{N}\sum_{k=1}^{N}\left(X^m - X^o\right)^2\right]^{\frac{1}{2}}, \tag{3}$$

$$\mathrm{S} = \frac{1}{2}\left(1 - \left|\frac{BIAS}{ABSE}\right| + \left|\frac{\mathrm{ABSE}}{\mathrm{RMSE}}\right|\right), \tag{4}$$

where X^o and X^m values are the observed and modelled values, respectively. BIAS/ABSE is bounded between −1 and 1, its target value being 0, which indicates that there is no systematic overestimation nor underestimation by the model. ABSE/RMSE is bounded between 0 and 1, with values close to 1 indicating that the distribution of $\left(X^m - X^o\right)$ has a large tail towards extreme positive or negative values. The target value for S is 1, which indicates that the model is unbiased and that its errors do not display too extreme values. It is worth noting that this skill score is not sufficient to give by itself an indication of the model performance, as it does not include any evaluation of the magnitude of the model errors relative to observation – a multiplication of all the error terms $\left(X^m - X^o\right)$ by a constant factor will leave the skill factor S untouched. Therefore, in the rest of the study, the S skill factor will be used alongside the absolute value of the bias as two indicators representative of the model ability to reproduce observations.

3 Results

3.1 Model results

Fig. 3 shows that the SO_2 and NO_2 concentrations simulated by the h25 simulation at the lowest model level are in excess of that simulated by the CTL simulation for all the simulated domain. This shows that the effect of injecting industrial emissions lower into the atmosphere has the effect of increasing simulated ground concentration of these primary species. As it could be expected, this effect is strongest where industrial emissions are the most important, namely in all the industrial Europe from Great Britain to Poland in northern Europe, and for particular regions such as the Po Valley in southern Europe. On the contrary, regions where the high NO_2 and SO_2 concentrations are due to sources other than stack emissions do not see a significant increase in the simulated concentrations from the CTL run to the h25 run (Fig. 1). This is for example the case of the areas of the Mediterranean Sea and North Sea that are affected by emissions related to navigation, but also of major urban areas such as Paris or London that are mostly affected by traffic and residential emissions.

Over the industrial regions, the year-average difference in SO_2 concentrations reaches 16 µg m^{-3} for areas of eastern Europe, which is more than doubling the SO_2 average concentration for these areas, reflecting the fact that the SO_2 background concentrations simulated in these regions are considerably higher in the h25 simulation than in the CTL simulation. The results for NO_2 are very similar, with simulated concentrations stronger in the h25 simulation than in the CTL simulation for the whole domain, with substantial differences over the major industrial regions of Europe, particularly in the eastern part of the continent.

3.2 Comparison to observations

3.2.1 Statistical comparison

The comparison of the CTL and h25 simulations to Airbase station data shows that the h25 simulation performs significantly better than the CTL simulation for the simulation of SO_2 concentrations (Table 3), particularly for the stations categorized as “rural background” in Airbase: for these stations, the average SO_2 concentration simulated by the h25 run is 2.6 µg m^{-3} (1.5 µg m^{-3} for the CTL run, 3.0 µg m^{-3} for the airbase measurements), so that the underprediction by CHIMERE has been reduced by 73 % by the change in the emission height. For other station types and locations, the simulated

SO_2 concentrations are increased in a comparable proportion between the CTL and h25 run, but the coarse resolution of these simulations does not allow the model to simulate correctly the impact of a highway, factory or urban area, therefore the simulated SO_2 concentrations are not comparable to observations for stations that are not "rural background" type. However, the model bias and skill scores are improved for all station types in spite of the lack of representation of local phenomena, due to the improved simulation of the background levels.

For NO_2, similar results are obtained when switching from the CTL to the h25 simulation, with enhanced simulation of the NO_2 background concentrations (reduction of the model bias by 66 % for the rural background stations, smaller reduction of the model bias for all station types). For O_3, all simulations exhibit excessive O_3 concentrations when compared to station measurements, which is an already known characteristic of the CHIMERE model, attributed to problems in modelling O_3 titration by NO (Solazzo et al., 2012). Due to redistribution of NO_x emissions closer to the surface in the perturbed simulations, a certain degree of improvement is obtained in these simulations. For the rural background stations, the annual bias in O_3 is reduced from $12.5\,\mu g\,m^{-3}$ to $10.6\,\mu g\,m^{-3}$, a 15 % reduction. A similar reduction is observed for all station types (about $1.8\,\mu g\,m^{-3}$ for the all-station average).

Therefore, as it is visible in Table 3 that a meaningful comparison between CHIMERE outputs and station data in the current CHIMERE configuration is possible only for rural background stations, we will examine below in more datail statistical indicators for the 94 selected stations only. For these 94 stations, a detailed examination of the model performances relative to observations according to 4 indicators (RMSE, AMSE, MB and Mao et al., 2006 skill score) permits to draw several conclusions as regards the compared skill of the 5 performed simulations (Table 4).

- The CTL simulation is always the most heavily biased of the 5 simulations, for O_3, SO_2 and NO_2. The concentrations of primary pollutants close to the surface tends to be systematically underestimated, and the O_3 concentration overestimated. The biases are reduced when going from CTL to h75, h50 and finally h25, which is the less biased simulation for the three examined species. Due to the smaller biases in h25, this simulation also has the best S score. From this point of view, the h25 simulation can be considered as the best choice among all 5 simulations, the CTL simulation being the worst choice.

- The error indicators RMSE and ABSE are systematically stronger in the h25 simulation for NO_2 and SO_2 in spite of the lower model bias, due to stronger model variability. The other four simulations yield rather similar ABSE and RMSE values, with the best values obtained in the Bie simulation for SO_2 and in the CTL and h75 simulations for NO_2. This indicates that higher variability in the h25 simulation may generate larger errors than in the other, more conservative, options
- Regarding O_3, the best simulation is h25 for all criteria, reducing the general model bias and the errors, and increasing the skill score. This is related to the fact that the h25 simulation has the strongest NO_x emissions in the lowest model levels, therefore increasing O_3 titration and tending to reduce the traditional high-ozone bias of CHIMERE (Solazzo et al., 2012). Regarding O_3, the worst simulation, with strongest biases and errors, is the CTL simulation, followed by the h75 simulation, the other two simulations (h50 and Bie) behaving rather similarly, with statistical indicators closer to these of h25 than of CTL.

The examination of these statistical indicators leads to several conclusions:

- The CTL simulation (emissions following the standard EMEP recommendations) seems to be clearly a bad choice for a wide range of criteria.
- h25 and Bie are two reasonable choices for the vertical distribution of emissions. h25 tends to reduce strongly the model biases, while generating larger errors for primary pollutants associated to larger model variability. The Bie simulation has good performances and is a more conservative choice than h25, with performance close to the best of the ensemble for all criteria. It also has the advantage to be

physically based on the plume-rise simulations of Bieser et al., 2011, while the h25 simulation is an arbitrary modification of the EMEP classical profiles, with only empirical basis.

3.2.2 Individual stations

Two individual stations have been selected to examine the time series of the h25, CTL and Bie simulations compared to the observations. DENW081 is a rural background station located in northwestern Germany, close to the border of the Netherlands. PL0243 is also a rural background station, located in southern Poland. These stations are interesting because they are located in regions in which heavy industrial influence occurs, but without being themselves close to a particular source, therefore representing a large-scale state for the industrial regions of the Ruhr and Silesia. As the h50 and h75 are always in-between the CTL and h25 simulation, their outputs are not presented in that figure. The first striking result here is that all three CHIMERE simulations behave similarly for the considered period and display a reasonable behaviour when compared to Airbase observations, both for DEN081 (Fig. 4) and PL0243 (Fig. 5). For both stations, as it is the case for the entire domain, the h25, Bie and CTL simulations are ordered by increasing concentrations of NO_2 and SO_2 and decreasing O_3. However, these differences have a different impact on the model's performance relative to the measurement station for these two locations.

For DEN081 (Fig. 4), all three simulations tend to slightly underestimate NO_2 and SO_2 peaks. The representation of the O_3 maxima is good, but the diurnal cycle of O_3 seems to be insufficient in this simulation, resulting in an overestimation of the average O_3 concentration. The differences between the simulations are as observed for the whole set of Airbase stations, with higher SO_2 concentrations in h25 during the whole period, which tends to be slightly closer to observed values when compared to the Bie simulation and the CTL simulation. SO_2 peaks are also enhanced in the h25 simulation and also, to a lesser extent, in the Bieser simulation, when compared to the Bieser simulation. O_3 differences between the three simulations are moderate, but quite measurable at night, where the nocturnal O_3 minimum values are more realistic in the h25 simulation than in the CTL or Bie simulations, particularly for periods with high NO_2 concentrations, when the NO_2 peaks and corresponding O_3 minima are represented better in the CTL simulation (see day 103 for example). The representation of O_3 maxima is rather good in all three simulations, and not considerably altered by the change in the vertical distribution of the emissions. All in all, for this particular station, the h25 simulation gives better results than the Bie simulation, which itself gives better results than the CTL simulation. The reduction in average O_3 concentrations is essentially due to O_3 titration at night by primary contaminants and does not necessarily imply a diminution of the daily maxima.

For PL0243 (Fig. 5), the SO_2 concentrations are either overestimated or underestimated by all three simulations simultaneously, with the differences reaching $80\,\mu g\,m^{-3}$ between the CTL and the h25 simulation, $30\,\mu g\,m^{-3}$ between the CTL and Bie simulation. Some peaks are adequately catched by the model, particularly in the Bieser simulation (days 315 and 325) or the h25 simulation (days 329-330). Generally, every time that a peak simulated in CHIMERE coincides with an observed peak, the peak is underestimated in the CTL simulation, which is not the case in the h25 or Bie simulations. Regarding NO_2, for this station, CHIMERE tends to overestimate the NO_2 concentrations for the considered period. This overestimation is weakest in the CTL simulation and strongest in the h25 simulation, even though the quantitative differences between these three simulations are not as strong as in the case of SO_2. In spite of this general overestimation, the agreement between simulated and measured NO_2 is globally good for this period, even though two significant peaks are simulated and not observed (around day 312 and day 342–343). A period of generally high NO_2 concentrations between day 325 and day 340 is adequately represented in all three simulations. However, for the whole considered period, NO_2 concentrations are slightly overestimated in all three simulations, more particularly in the Bie and h25 simulations. Regarding the O_3 concentrations, their representation is good in all three simulations, both O_3 peaks and minima tend to be represented correctly. As could be expected, the higher NO_2 concentrations in the h25 and Bie runs are associated to more pronounced O_3 minima. Compared to the DENW081 station, the more realistic representation of NO_2 concentration and O_3 concentration minima relative to Airbase seems to indicate a better representation of the NO-NO_2-O_3 cycle for this grid cell, either due to better emissions or to the meteorological conditions. It is also arguable that the NO_2 concentrations measured in DENW08 are not representative of the entire grid cell, since the DENW081 Airbase station is located in the town of Borken (Germany), with possible local traffic effects, unlike Potok Zloty (Poland) which is just a village.

4 Discussion and conclusions

Five air quality simulations have been performed with CHIMERE from 20 February 2008 to 19 February 2009 for a domain covering Europe at 0.5° horizontal resolution, with a meteorological simulation from WRF and anthropogenic emissions from the EMEP database at 0.5° resolution. These five simulations have been conducted using either the EMEP recommandations for vertical disaggregation (CTL simulation), modifying them by a factor of 0.25, 0.50 and 0.75 (h25, h50 and h75 simulations respectively), or using the alternative profiles of Bieser et al., 2011 (Bie simulation). The vertical emission profiles were the only difference between these 5 simulations, which have been performed using the same meteorology, the same horizontal and temporal repartition of the emissions, and the same CTM with the same configuration. Therefore, their results could be analyzed directly in terms of the impact of vertical disaggregation factors on simulated pollutant concentrations.

Due to the relatively coarse model resolution, the model results are representative mainly of the concentrations as measured by the rural background stations. For these stations, the h25 simulation permits a reduction of the model bias of respectively 73 %, 66 % and 15 % for SO_2, NO_2 and O_3, so that in the case of SO_2 and NO_2, the errors on simulated concentrations due to the uncertainties on the effective emission heights have the same order of magnitude than the biases of the simulated concentrations relative to ground measurements, so that improving the evaluation of the effective emission height is a key factor in improving the representation of the atmospheric composition. The interpretation of the large bias reduction obtained when reducing the effective emission heights for stack emissions by switching from the standard EMEP recommendations to the Bieser et al., 2011 recommendations is the same for SO_2 and NO_2 which are primary pollutants. For these pollutants, the reduction in the model underprediction can be directly attributed to the fact that industrial NO_2 and SO_2 is emitted at lower model levels in the h25 simulation than in the CTL simulation. For O_3, which is not a primary pollutant, the reduction in CHIMERE overprediction could be a consequence of stronger simulated NO_x concentrations in the lowest atmospheric layers, leading to stronger O_3 titration by NO. The h75 and h50 simulations display characteristics that are intermediate between h25 and CTL, improving the CTL simulation relative to Airbase measurements. The simulation conducted using the Bieser et al., 2011 profiles displays a behaviour close to the h50 simulation, with also a notable improvement compared to the CTL simulation, and notable bias reduction regarding NO_2, SO_2 and O_3 concentrations. Comparison to station data shows that the impact of the different vertical emission profiles on NO_2 and SO_2 concentrations can affect the simulated SO_2 concentrations by a factor 2, including for peak values, and even for rural background stations, so that the impact of vertical emission profiles on the modelling of SO_2 is fundamental, and much attention should be devoted to this problem. For NO_2, the impact is quite significant too, with the use of the h25 or Bieser vertical profiles contributing to reducing the low bias of CHIMERE simulated NO_2 concentrations for stations where CHIMERE exhibits this low bias, which is the case of most stations. In such cases, as for the DENW081 Airbase station, lowering the vertical emission profiles has the effect of reducing the bias and therefore indirectly improving the simulation of O_3 concentrations, particularly nightime O_3 titration.

These results show that, due to very large incertainties in its computation, vertical disaggregation can be a major error cause in air quality modelling particularly in the regions influenced by large stack emissions such as large parts of eastern Europe and the most industrial parts of western and southern Europe. Overestimation of effective emission heights, as seems to be the case of the EMEP recommandations, is a determinant contribution to NO_2 and SO_2 underestimation in CHIMERE when using the EMEP emission dataset, and to O_3 overestimation, particularly suring nighttime. These biases can be corrected in a relatively straightforward way by applying alternative vertical emission profiles that lead to lower effective emission altitude, such as the ones proposed by Bieser et al., 2011. Therefore, the use of the Bieser et al., 2011 profiles will be proposed in future versions of CHIMERE, which should be a way of generally improving the simulated concentrations of primary and secondary contaminants of anthropic origin. As this study has been performed with only one model, CHIMERE in its 2011 version, and one vertical discretization, its results might not be directly applicable to other CTMs or other vertical resolutions. However, the relatively coarse vertical resolution used in this study is a factor that can increase numerical diffusion on the vertical and therefore reduce the impact of the vertical distribution of emissions, so that it can be thought that the effect of the uncertainties on effective emission heights on the modelling of atmospheric composition will be even stronger for model configurations with more vertical layers, inducing less vertical mixing.

The authors think that these questions should receive increased attention in the following years, due to their strong impact on the simulated concentrations of all chemical species influenced by industrial activities. As shown here for CHIMERE, a reevaluation of the vertical emission heights using state-of-the-art vertical profiles instead of profiles that have been provided using earlier methodologies might bring significant added value to the simulated concentrations for other CTMs as well.

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Table 1. (Top) Vertical disaggregation factors per EMEP levels (left column) and per SNAP sectors as recommended in EMEP with additionnal 0–20 m layer for surface emissions, as used by default in CHIMERE, (Bottom) Vertical disaggregation factors per EMEP levels (left column) and per SNAP sectors from Bieser et al., 2011, with additionnal 0–20 m layer for surface emissions.

	SNAP sectors									
EMEP layers	1	2	3	4	5	6	7	8	9	10
Default vertical disaggregation										
0–20	0.0	11.0	0.0	20.0	20.0	100.0	100.0	100.0	2.0	100.0
20–92	0.0	39.0	0.0	70.0	70.0	0.0	0.0	0.0	8.0	0.0
92–184	0.0	50.0	4.0	10.0	10.0	0.0	0.0	0.0	15.	0.0
184–324	8.0	0.0	19.	0.0	0.0	0.0	0.0	0.0	40.	0.0
324–522	46.	0.0	41.	0.0	0.0	0.0	0.0	0.0	35.	0.0
522–781	29.	0.0	30.	0.0	0.0	0.0	0.0	0.0	0.0	0.0
781–1106	17.	0.0	6.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Vertical disaggregation modified after Bieser et al., 2011,										
0–20	0.0	11.0	0.0	20.0	20.0	100.0	100.0	100.0	2.0	100.0
20–92	0.0	89.0	21.3	70.0	70.0	0.0	0.0	0.0	8.0	0.0
92–184	0.25	0.0	75.4	7.0	6.0	0.0	0.0	0.0	37.	0.0
184–324	51.0	0.0	3.3	1.0	3.0	0.0	0.0	0.0	51.	0.0
324–522	45.3.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	2.0	0.0
522–781	3.29	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
781–1106	0.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0

Table 2. Year-average vertical repartition of SO_2 emissions (%) for the five simulations that have been performed.

Simulation CHIMERE layer	CTL	Bie	h75	h50	h25
0–25	29.3	29.8	30.1	31.8	36.1
25–71	4.4	8.4	5.7	6.4	8.6.
71–158	5.5	10.8	6.0	8.0	35.8
158–321	7.7	26.0	18.2	34.9	19.5
321–532	33.6	24.0	32.3	18.9	0.0
532–1240	19.5	1.1	7.7	0.0	0.0
1240–2493	0.1	0.0	0.0	0.0	0.0

Table 3. Average SO_2, NO_2 and O_3 concentrations ($\mu g\,m^{-3}$) for all classes of stations for the CTL simulation (before / character) and the h25 simulation (after / character). In brackets is the average Airbase value for the considered category of stations. In italics are the categories that include less than 10 stations. The stations are classified according to the Airbase reported zone (Rural/Suburban/Urban/Indeterminate) and type (Background/Traffic/Industrial/Indeterminate) of the station.

	BG	INDUS	TRAF	INDET	ALL
SO_2					
RUR	1.5/2.7(3.0)	1.7/2.8(5.0)	2.0/3.7 (5.2)	–	1.8/3.2 (4.6)
SUB	1.2/1.6 (2.9)	1.7/2.8 (5.9)	2.5/4.8 (11.3)	–	1.9 3.3 (7.3)
URB	–	1.6/2.7 (5.9)	1.6/2.4 (6.0)	–	1.6/2.5 (6.0)
INDET	1.4/2.9 (4.0)	1.6/2.2 (5.9)	1.3/1.7 (6.1)	*4.8 5.3 (4.5)*	1.5/2.3 (5.4)
ALL	1.4/2.7 (3.3)	1.7/2.6 (5.5)	1.8/3.0 (5.9)	*4.8 5.3 (4.5)*	1.7 2.9 (5.3)
NO_2					
RUR	8.4/10.0(10.3)	9.3/10.7 (23.0)	10.9/12.8 (26.6)	–	9.9/11.6 (21.4)
SUB	5.5/6.2 (12.7)	7.2/8.4 (17.1)	9.5/11.7 (24.1)	–	7.7/9.2 (18.8)
URB	–	7.7/9.7 (28.2)	9.7/11.0 (37.2)	–	9.4/10.8 (36.0)
INDET	5.1/6.3 (8.3)	7.8/9.0 (18.5)	8.3/9.4 (25.0)	*23.9 26.2 (41.1)*	7.3/8.5 (17.9)
ALL	7.2/8.6 (9.8)	8.4/9.7 (21.2)	10.0/11.6 (29.6)	*23.9 26.2 (41.1)*	9.0/10.2 (23.2)
O_3					
RUR	73.1/71.2 (60.6)	76.6/74.9 (49.0)	70.3/68.2 (44.9)	–	72.5/70.6 (50.0)
SUB	88.8/87.8 (57.3)	82.6/81.2 (50.9)	78.0/75.6 (54.3)	–	82.0/80.3 (53.0)
URB	–	81.3/79.2 (41.3)	80.9/79.2 (41.4)	–	80.9/79.2 (41.4)
INDET	86.3/84.6 (63.4)	84.7/83.1 (53.6)	82.6/81.0 (49.1)	*72.9 70.2 (32.3)*	84.4/82.8 (54.8)
ALL	78.3/76.6 (61.4)	80.5/78.9 (50.0)	76.0/74.1 (44.9)	*72.9 70.2 (32.3)*	77.7/75.9 (49.7)

Table 4. RMSE, ABSE, BIAS and S (skill score) for SO_2, NO_2 and O_3, averaged for the 94 selected rural background stations, for each of the 5 performed simulations. For each indicator, the best value(s) is (are) in bold font, the worst value(s) in italic font.

	RMSE	ABSE	BIAS	S
SO_2				
h25	*4.03*	*2.46*	**–0.34**	**0.55**
h50	3.78	2.28	–0.93	**0.55**
h75	**3.75**	2.26	–1.24	0.52
CTL	3.76	2.28	*–1.48*	*0.50*
Bie	**3.75**	**2.25**	–1.00	0.54
NO_2				
h25	*10.3*	*7.17*	**–0.29**	**0.62**
h50	10.1	6.96	–1.12	**0.62**
h75	**10.0**	6.92	–1.55	0.61
CTL	**10.0**	**6.91**	*–1.88*	*0.60*
Bie	10.1	6.96	–1.11	**0.62**
O_3				
h25	**27.1**	**21.8**	**10.6**	**0.64**
h50	27.2	22.0	11.4	0.63
h75	27.4	22.2	12.0	*0.62*
CTL	*27.5*	*22.3*	*12.5*	*0.62*
Bie	27.2	22.0	11.4	0.63

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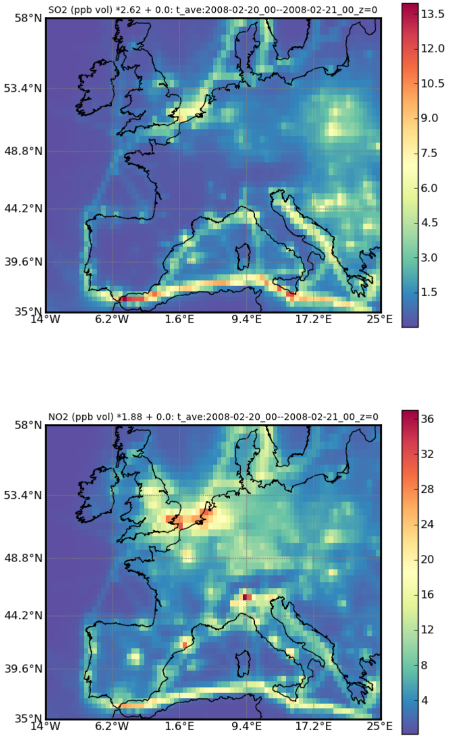

Fig. 1. (Top) average SO_2 concentration simulated at the first model level in the CTL simulation ($\mu g\,m^{-3}$), (Bottom), same as (Top) but for the NO_2 concentration.

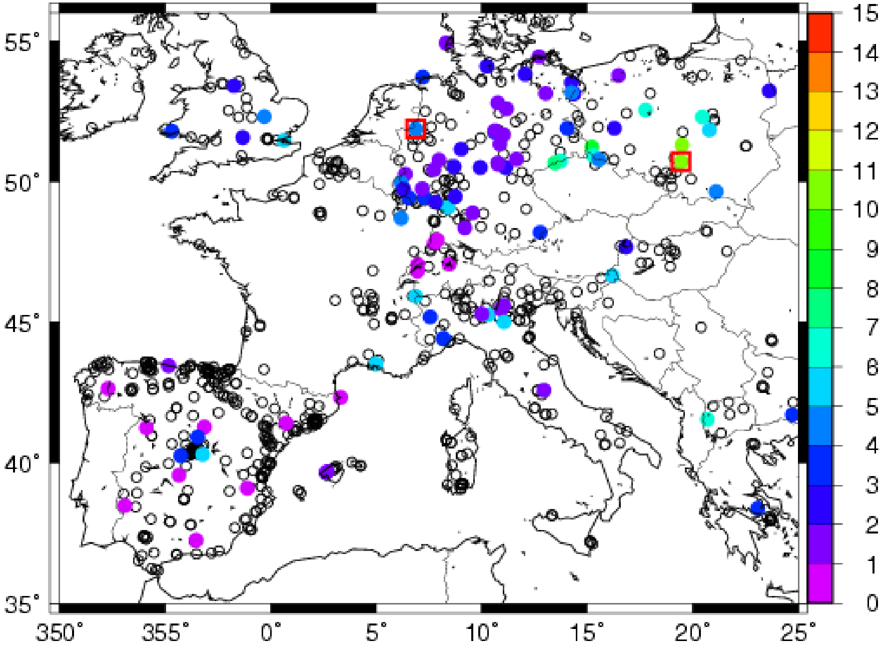

Fig. 2. Map of the 721 Airbase stations used in the study (circles), with average SO_2 concentration ($\mu g\,m^{-3}$) from 20 February 2008 to 19 February 2009) in color for the 94 stations classified as Rural background. Stations DENWO81 (close to the German-Dutch border) and PL0243 (center-south Poland) studied in more detail are enclosed within a red square

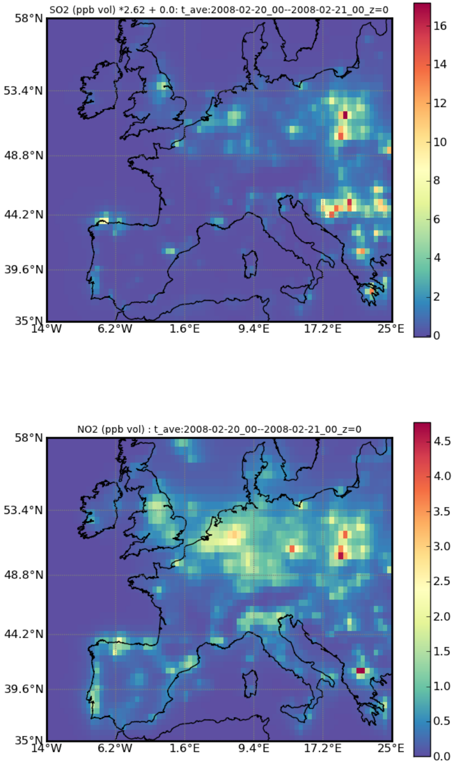

Fig. 3. (Top) year-averaged difference between the year-average SO_2 concentration in the h25 and CTL simulation (µg m^{-3}) and (Bottom) same for the NO_2 concentrations.

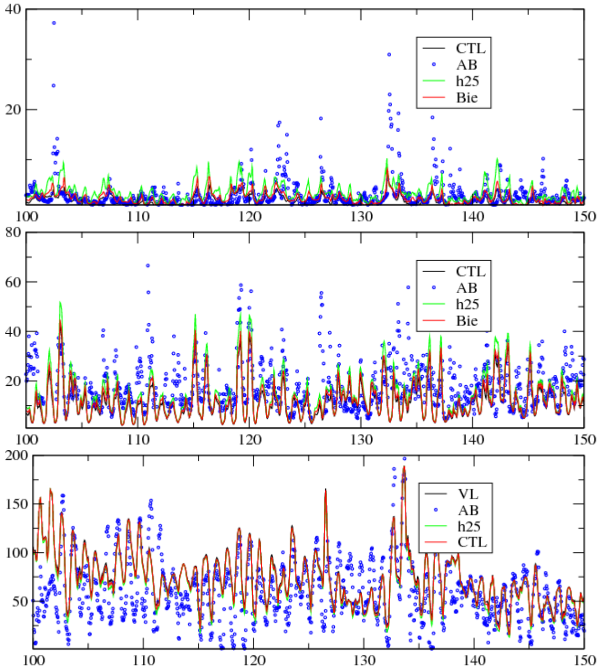

Fig. 4. Time series for Airbase station DENW081 (Borken-Gemen, 51.86° N–6.87° E), rural station in North-Westphalen, from 29 May 2008 to 18 July 2008. Time is in days from the start of the simulation, time series for SO_2 (Top), NO_2 (Middle) and O_3 (Bottom) are presented, in $\mu g\,m^{-3}$.

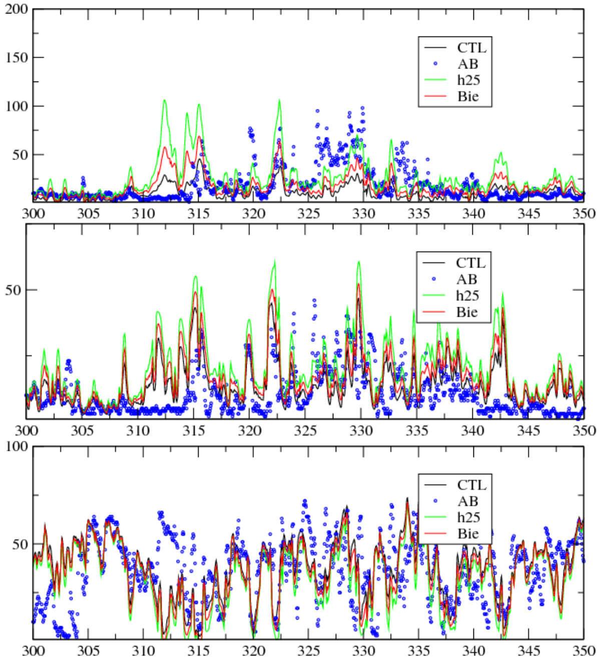

Fig. 5. Time series for Airbase station PL0243 (Potok-Zloty, 50.71° N–19.46° E), rural station in Lower Silesia (Poland), from 15 December 2008 to 3 February 2009. Time is in days from the start of the simulation, time series for SO_2 (Top), NO_2 (Middle) and O_3 (Bottom) are presented, in $\mu g\,m^{-3}$.