Geosci. Model Dev. Discuss., 6, 4189–4205, 2013
www.geosci-model-dev-discuss.net/6/4189/2013/
doi:10.5194/gmdd-6-4189-2013

Geoscientific Model Development Discussions
Open Access

Frontiers in air quality modelling

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Received: 8 July 2013 – Accepted: 17 July 2013 – Published: 2 August 2013

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Published by Copernicus Publications on behalf of the European Geosciences Union.

Abstract

The first pan-European kilometre-scale atmospheric chemistry simulation is introduced. The continental-scale air pollution episode of January 2009 is modelled with the CHIMERE offline chemistry-transport model with a massive grid of 2 million horizontal points, performed on 2000 CPU of a high performance computing system hosted by the Research and Technology Computing Center at the French Alternative Energies and Atomic Energy Commission (CCRT/CEA). Besides the technical challenge, we find that model biases are significantly reduced, especially over urban areas. The high resolution grid also allows revisiting the contribution of individual city plumes to the European burden of pollution, providing new insights for designing air pollution control strategies.

1 Introduction

Beside the challenges in understanding and representing physical and chemical processes, the most common limitation mentioned in geophysical modelling studies regards spatial resolution, often restricted because of computational cost. With the ever growing computing facilities, such limitations are systematically pushed back and we are now able to introduce a continental scale atmospheric chemistry simulation at 2 km resolution (whereas such high spatial resolutions are limited to local air quality models, Zhang et al., 2012). This paper puts in perspective this achievement with the current state of the art in regional atmospheric chemistry modelling. We highlight the strength and weaknesses of the simulation and discuss the relevance of very high performance computing in the field of environmental modelling.

While the air quality situation for nitrogen dioxide (NO_2) and fine particulate matter ($PM_{2.5}$) improved over the past decade (EEA, 2011), the situation is still far to match the guidelines of the World Health Organisation. In 2009, the European annual limit value

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was exceeded for NO_2 at 41 % of traffic stations and about 20 % of the European urban population was exposed to PM_{10} pollution above the daily limit value (EEA, 2011).

One of the main air pollution outbreak in Europe in 2009 occurred between 9 and 16 January with particulate and nitrogen dioxide levels reaching alert levels (MEEDDM, 2010; Chiappini, 2009; Favez et al., 2010) in various places. The main driving factor for this event was an intense cold spell (DWD, 2010) that led to high anthropogenic emissions of pollutants from domestic heating and a stable meteorological situation enhancing the accumulation of air pollution in the thin planetary boundary layer. While Western Europe was exposed to high levels of pollution at the beginning of the period, the pollution plume moved eastwards towards the end of the period as a result of the displacement of the cold spell and the advection of western pollution. The conjunction of long-range transport and contribution of local sources supports the relevance of this event to investigate the added value of high-resolution continental-scale atmospheric chemistry simulations. The choice of a wintertime event to assess the added value of spatial resolution is further supported by previous studies that, by comparing 36 and 4 km simulations over the Paris area, demonstrated the lower sensitivity in summer, when the formation of secondary pollutant smear out the spatial variability (Fountoukis et al., 2013).

The horizontal resolution in the simulation proposed here is about 2 km, the lower limit being set by the availability of proxy data in building the emission inventories rather than computational capacities or cost. With such a resolution, the European continent is covered with slightly more than 2 million horizontal grid cells. These numbers should be put in perspective with current practices in air pollution forecasting, where a resolution of about 10 km is achived by the Modelling Atmospheric Composition Change project (MACC) of the Global Monitoring for Environment and Security programme of the European Commission (www.gmes-atmosphere.eu, Zyryanov et al., 2011) and the US air quality forecasting system of the National Oceanic and Atmospheric Administration and Environmental Protection Agency (Eder et al., 2009). The computational demand of the simulation presented here is thus two orders of magnitude (5 × 5 for the

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number of horizontal points, and another factor 5 for the increment in the time step) above current practices.

2 Methods

The offline Chemistry Transport Model (CTM) CHIMERE (Menut et al., 2013) was used to model the transport and transformation of trace species in the lower troposphere. The model is being used by several international teams for research and environmental policy underpinning. It has been involved in a number of model intercomparison initiatives (Colette et al., 2011; Cuvelier et al., 2007; van Loon et al., 2007; Vautard et al., 2009; Galmarini et al., 2012) and is now part of the European pre-operational air composition forecasting system MACC (Zyryanov et al., 2011) within the GMES program. The model and further documentation can be found at www.lmd.polytechnique.fr/chimere. The version used here is CHIMERE-2008 (Bessagnet et al., 2008) modified to include a better representation of turbulent mixing in urban areas as described in (Terrenoire et al., 2013) who use an identical model version and setup as here.

As an offline CTM, CHIMERE requires prescribed meteorological fields which were provided here by ECMWF with the IFS model at 16 km resolution with data assimilation. Although the resolution of the meteorological model does not match that of the CTM, performing a continental scale high-resolution meteorological simulation including urban island effects is a challenge in itself that was ruled out of the present initiative.

The efforts of the CHIMERE development team for the present initiative were focused on the improvement of the anthropogenic emissions module (Menut et al., 2012). A few years ago, frontiers in atmospheric chemistry simulation were set by the resolution of emission inventories that were delivered at approximately 50 km at continental scale, whereas higher resolution (up to 1 km) could only be reached over a restricted area with bottom-up inventories. However, because of heterogeneities in the input data used in bottom-up emission datasets and lack of required information at the local scale,

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such an approach cannot yet be contemplated at continental scale. Instead, we used a top-down approach, i.e. downscaling techniques to disaggregate coarse inventories using proxies such as point source location and high-resolution population density map. Here the resolution limitation is thus prescribed by the proxy data: in our case the population density map that was made available at a resolution of about 1 km by the Joint Research Centre of the European Commission.

More specifically, the anthropogenic emission used here is the inventory developed by INERIS in the framework of the EC4MACS project (European Consortium for Modelling of Air Pollution and Climate Strategies, http://www.ec4macs.eu/). It is described in further details in Bessagnet et al. (2012) and Terrenoire et al. (2013) and consists of an optimal combination of various sources:

- TNO $0.125^{\circ} \times 0.0625^{\circ}$ emissions for 2007 from the MACC project (Kuenen et al., 2011).
- EMEP officially reported national totals gridded at $0.5^{\circ} \times 0.5^{\circ}$ resolution for the year 2009 (Vestreng et al., 2007).
- Emission data from the GAINS database (Amann et al., 2011, http://gains.iiasa.ac.at/gains).

Industrial emissions provided in the EMEP inventory were spatially distributed at the location of large and medium size point sources from the EPER database of EEA (http://www.eea.europa.eu/data-and-maps/data/eper-the-european-pollutant-emission-register-4). Domestic emissions were spatially distributed using population density maps and correlation coefficient derived from a French bottom-up inventory (Bessagnet et al., 2012). Total domestic emissions are those of the EMEP inventory except for the following countries where GAINS estimates are used: CZ, BA,BE, BY, ES, FR, HR, IE, LT, LU, MD, MK, NL, CS, TR. Non-industrial and non-domestic emissions are those of the EMEP inventory, regridded using the TNO-MACC inventory as spatial proxy.

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A resolution of 0.03125 and 0.015625 degrees (i.e. about 2.5 and 1.5 km at the location of Paris) of longitude and latitude, respectively, was chosen. The horizontal geographical domain includes 1309 × 1661 points for 9 vertical layers. The simulations were parallelised on 2000 Intel Xeon processors at 2.7 GHz with 64 GB memory by node. Tests were performed with up to 4000 processors with less satisfactory scaling properties. The total cost of the project was about 110 000 h core, or 13 yr on a single processor. The storage of input/output files also constitutes an achievement with more than 2T of data. The simulation covers 9 days (8 January 2009 to 16 January 2009). Initial conditions are provided by the 7 km simulation of (Terrenoire et al., 2013) with one month spin up so that only the first day is considered as a spin up of the high-resolution simulation and discarded from the quantitative analysis below. The high resolution domain is nested in a coarser (50 km) CHIMERE simulation, itself using global boundary conditions from a 6 yr reanalysis with the LMDz4-INCA3 model (Folberth et al., 2006; Hauglustaine et al., 2004). The physical and chemical setup of the model is identical to (Terrenoire et al., 2013).

3 Results

The maps in Fig. 1 illustrate the added value of increased resolution by showing snapshots of the simulations of nitrogen dioxide at about 50 and 2 km of resolution. A more dynamic view is accessible in the movie provided in the Supplement. While the coarse run captures well the main emission hotspots, local maxima over individual roads and isolated point sources are smeared out because of the numerical diffusion of the Eulerian model.

A more quantitative analysis of the added value of resolution will be provided in the following paragraphs, but the benefit of such a high resolution simulation in raising awareness on the contribution of local sources in the accumulation of air pollution at the continental scale is worth mentioning. While discussions about the design of efficient air pollution mitigation policy measures often falls back to the choice of the most

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relevant geographical scale (local, national, continental), these high-resolution maps constitute a relevant reminder that in the field of atmospheric pollution as well, little brooks make great rivers.

A comparison with the observations allows discussing the improvement in the performance of the model. An average of time series of NO_2 at all urban monitoring stations in the Paris area is given in Fig. 2. The stations used to compute this composite time series are the regulatory monitoring sites of the European network compiled in the AIRBASE repository (eea.europa.eu/themes/air/airbase) included in the geographical box encompassing the larger Paris area: 47° N, 50° N, 1.5° W, 3.5° W. It illustrates well the improvement brought about by the increased resolution to capture high pollution levels in urban areas. Whereas the coarse simulation is only able to reproduce the diurnal cycle and the daily variability with an important average bias, moving to a 7 km resolution improves the average level and daytime values, while night-time and morning peaks are further improved when moving to a 2 km resolution.

The performances of the model are further quantified in Table 1 that synthesizes the average root mean square error (RMSE) across the whole AIRBASE network as well as daily spatial correlation coefficients averaged over the whole period. We find that increasing the resolution improves significantly the root mean square error, especially at urban sites. The difference between the RMSE at urban, suburban and rural stations decreases with the resolution showing that we resolve better spatial gradients. This feature is more sensitive for NO_2 than PM_{10} because of the lower spatial variability of the later yielding this higher sensitivity of NO_2 often reported at urban sites.

Table 1 somehow questions the relevance of very high resolution simulations since the bulk of the improvement is achieved at 7 km resolution for which the RMSE of daily mean NO_2 averaged over the whole monitoring network is 26 % (10 % for PM_{10}) lower than the 50 km simulation and the 2 km resolution adds only another 5 % (3 % for PM_{10}) reduction to achieve an average RMSE 32 % (13 % for PM_{10}) lower than the coarse configuration. In addition, the improvement in the spatial correlation is only seen at rural sites (and suburban sites for PM_{10}). These features should not overshadow the

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improvement in terms of net RMSE brought about by the increased resolution. However, one could have expected that the improvement could have been even larger. These figures illustrate the need to investigate further more sophisticated emission downscaling algorithms, and accounting for small-scale dynamical meteorological features (such as urban processes).

However, Table 1 only addresses average biases and similar limited added value of increased resolution were reported before as far as mean or fractional biases are concerned (Fountoukis et al., 2013; Tesche et al., 2006). On the contrary the performances in terms of exceedances (which are notably challenging to capture with numerical models because of thresholding effects and of high regulatory relevance in the European air quality legislation, EC, 2008) are improved at very high resolution. For PM_{10}, the daily limit value of 50 $\mu g\,m^{-3}$ was exceeded 1761 times throughout the European monitoring network between the 9th and 16th of January 2009. Using a 50 km resolution, only 10 % of the exceedances are captured by the CTM, but the increased resolution improves this number with 31 % and 50 % of the cases captured by the model at 7 km and 2 km of resolution, respectively.

The non-linearity of the atmospheric chemistry system yields a slight sensitivity to the resolution of the total mass of modelled particulate matter aggregated over Western Europe (10° W, 30° E, 36° N, 60° N). The average PM_{10} load is 17.4, 19.5 and 20.7 $\mu g\,m^{-3}$, for the 50, 7, and 2 km resolution simulations respectively, while the corresponding figures for NO_2 are 10.2, 13.8, 14.4 $\mu g\,m^{-3}$.

The increased resolution has thus also an impact on the net fluxes. We find that ignoring fine plumes in the coarse simulation leads to an underestimation of long range transport. In particular the eastward flux of $PM_{2.5}$ built up over Western Europe during the first half of the event is underestimated by 15 % in the 50 km simulation compared to the 2 km simulation. When focusing on individual cities, we found that the net export of $PM_{2.5}$ in the immediate vicinity of the Paris area (1.4375° E, 3.1875° E, 47.96875° N, 49.71875° N) is 36 % (resp. 22 %) higher in the 2 km (resp. 7 km) resolution simulation compared to the 50 km run. For NO_2, these figures are 48 % and 50 % for the 2 km and

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7 km resolution. It should be emphasised that total emission fluxes are identical in all simulations. The differences obtained for the export flux of NO_2 are thus partly induced by the difference in the spatialisation of these emissions that are smeared out in the coarse simulation over the boundaries of the Paris area defined above. The similarity of export flux for NO_2 in the 2 km and 7 km simulation shows that the intermediate 7 km setup is somewhat satisfactory to capture urban NO_2 patterns, considering available emission data. On the contrary, we find an additional improvement at 2 km for $PM_{2.5}$ attributed to the non-linear chemical formation of secondary particles.

The sensitivity is even much larger when it comes to exposure modelling. Because highly populated areas are correlated with emissions hotspots, the horizontal gradients matter when weighting pollution load by population density. Here we find that on average over the 9 days period, in the high resolution simulation 87.9 million people (out of 588 million inhabitants included in the modelling domain) were exposed to daily concentrations exceeding the daily limit value of 50 $\mu g\,m^{-3}$ for PM_{10} according to the EU Directive on the ambient air quality (2008/50/EC). Using a 50 km grid, the same estimate would be 18.2 million people, i.e. almost a factor 5 underestimation if the additional increment of pollution in urban areas is ignored.

4 Conclusion

By producing the first continental-scale 2 km resolution air quality simulation performed on a 2000 CPU computer, we introduced the first steps of atmospheric chemistry modelling in the era of very high performance computing. This frontier simulation is two orders of magnitude above current practices in terms of computational demand.

Besides the technical challenge, which demonstrated the robustness of the selected air quality model, we discuss the added value in terms of air pollution modelling and decision support. The comparison with in-situ observations shows that model biases are significantly improved despite some spurious added spatial variability attributed

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to shortcomings in the emission downscaling process and coarse resolution of the meteorological fields.

The increased spatial resolution is clearly beneficial for the detection of exceedances, the quantification of transport fluxes and exposure modelling resulting from the non-linearity of the atmospheric system. We exhibit dynamic air pollution patterns that highlight the contribution of the city plumes to the background air pollution levels. Reciprocally, the contribution of long range transport to exceedances of regulatory threshold values for PM_{10} and NO_2 in urban areas is better assessed as a result of reduced diffusion. The underestimation in terms of net outgoing flux of trace species is of the order of 15 %, which raises important concerns in studies focusing on impacts on remote areas with large scale coarse models. Lastly, up to a factor 5 underestimation of the fraction of population exposed to detrimental levels of pollution can be derived with a simulation at coarse resolution if subgrid scale correction such as urban increments (Amann et al., 2011; Denby et al., 2011) are ignored to cope with the lack of explicit high resolution model.

We conclude that significant improvement can be sought after by refining the geographical mesh of atmospheric chemistry models. Better results can be achieved thanks to a better representation of the contribution of local sources in the build-up of continental scale events, in the net transport fluxes and in the aggregated population exposure to detrimental air pollution levels. This experiment opens new perspectives for decision making. In Europe, considering the achievements after two decades of efforts to reduce air pollutant emissions the challenge is now to find the optimal trade-off between national and local air quality management strategies. While the first approach is based on sectoral strategies and energy policies, the later builds upon new alternatives such as urban development. The strategies, the decision pathways and the involvement of individual citizen differ, and a compromise based on cost and efficiency must be found. We illustrated how high performance computing in atmospheric science can contribute to this aim. Although further developments are still needed to secure the results for routine policy use.

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Supplementary material related to this article is available online at: http://www.geosci-model-dev-discuss.net/6/4189/2013/gmdd-6-4189-2013-supplement.zip.

Acknowledgements. The high performance computing facility and staff at CCRT/CEA is gratefully acknowledged. EEA granted access to the AIRBASE monitoring data and ECMWF to meteorological forcing. European funding also benefited to this initiative through the MACC-II (grant agreement no 283576) project of the European Union's Seventh Framework Programme (FP7/2007–2013) and the EC4MACS project of the Life Programme of the European Commission (LIFE06 ENV/AT/PREP/06).

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Table 1. Model performances (root mean square error and average spatial correlation) of each configuration over the whole modelling domain between the 9 and 16 January 2009.

Pollutant	Station Type	50 km		7 km		2 km	
		RMSE ($\mu g\,m^{-3}$)	Corr.	RMSE ($\mu g\,m^{-3}$)	Corr.	RMSE ($\mu g\,m^{-3}$)	Corr.
NO_2	Urban	29.95	0.70	21.52	0.65	18.93	0.62
	Suburban	17.03	0.60	12.30	0.58	11.77	0.56
	Rural	7.80	0.54	6.67	0.64	6.68	0.68
PM_{10}	Urban	34.14	0.57	31.28	0.46	30.42	0.43
	Suburban	17.55	0.49	15.42	0.51	14.54	0.53
	Rural	6.96	0.51	6.26	0.63	6.01	0.63

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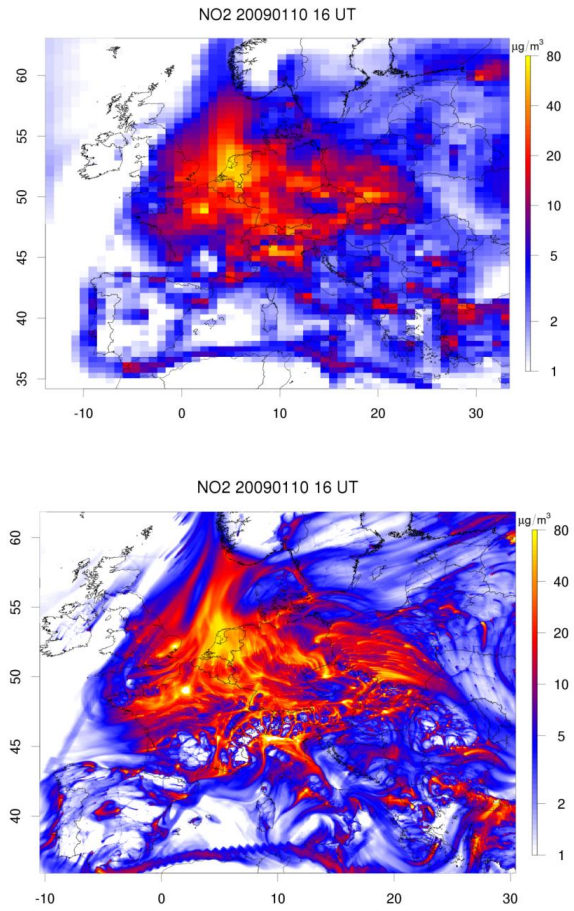

Fig. 1. Map of NO_2 ($\mu g\,m^{-3}$) on 10 January 2009 at 16:00 UTC in the 50 km (top) and 2 km (bottom) resolution CHIMERE simulations.

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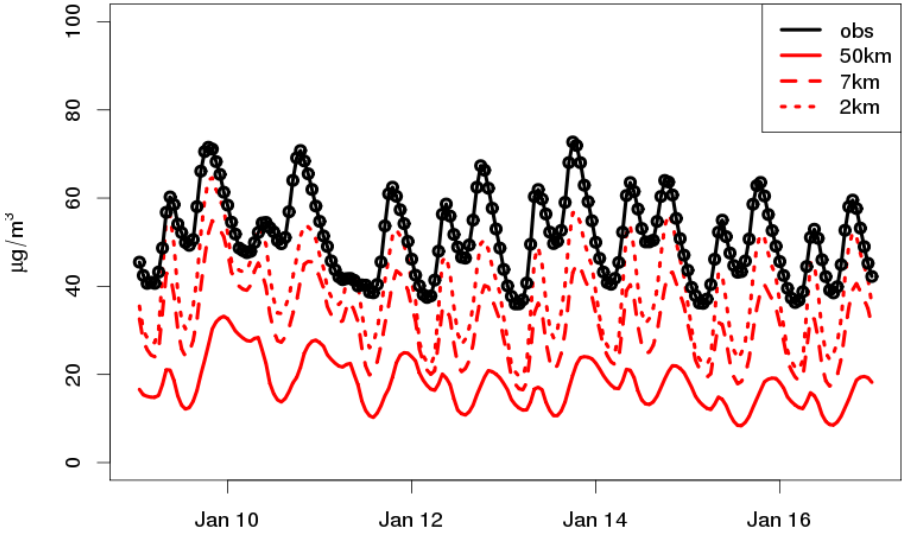

Fig. 2. Average time series of hourly NO_2 ($\mu g\,m^{-3}$) observed at urban background monitoring stations in the Paris area (black) and interpolated in the CHIMERE model simulations at the 50 km (solid), 7 km (dashed), and 2 km (dotted) resolution.

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