Air Qual Atmos Health
DOI 10.1007/s11869-015-0315-x

Modelled NO_2 tropospheric columns at different resolutions versus OMI satellite data: analysis of a 1-year BOLCHEM simulation over Europe

Lakhdar Aidaoui · Alberto Maurizi · Abbes Azzi

Received: 11 July 2014 / Accepted: 7 January 2015

Abstract Model simulations of tropospheric NO_2 vertical column density are performed using the online-coupled BOLCHEM model. Model output is compared to ozone monitoring instrument (OMI) data from the Tropospheric Emission Monitoring Internet Service (TEMIS) over Europe for the year 2007. European hot-spots (Po Valley and BeNeLux) are simulated at finer resolution and analysed separately, along with the area of Gibraltar. Standard statistical analysis reveals good model performances, even in highly polluted regions, with spatial correlation 0.90 for the whole of Europe, 0.74 for the Po Valley, 0.85 for BeNeLux and 0.79 for Gibraltar. Seasonal analysis shows some dependency on time, with lowest scores in winter, when the satellite product also suffers weaker statistical significance due to the presence of clouds. The increase in resolution is found to affect the spatial correlation more the Po Valley (+23 %) than in BeNeLux (+5 %). This difference is likely to depend on the very different meteorology of the two hot-spots.

Keywords Tropospheric NO_2 column · Emission hot spots · BOLCHEM model · Model resolution effect · OMI sensor

Introduction

Evaluation of the spatial and temporal distribution of Nitrogen dioxide (NO_2) concentration in the troposphere has attracted much interest from the scientific community, from both monitoring and modelling points of view. NO_2 is one of the most important atmospheric pollutants due to its effect on human health (see, e.g. Latza et al. 2009), and, specifically, its influence on mortality (see, e.g. Chen et al. 2012. Furthermore, it plays a basic role in the formation of ground ozone which is know to be harmful, not only for human, but also for ecological health (Fuhrer et al. 1997; Ashmore 2005). As a consequence, it is one of the few pollutants that are regulated by the environmental policy and, accordingly, it is considered to be one of the main indexes of local pollution (Richter et al. 2005; Monks et al. 2009). It also affects the climate by increasing the levels of greenhouse gases (Solomon et al. 1999). A small fraction of NO_2 is directly emitted while the largest part is a secondary product and derives basically from emitted NO. NO + NO_2 constitutes the NOx family. NOx originates from different sources (combustion of fossil fuel, biomass burning, microbiological processes in soil, lightning, wildfires, etc.) but it is mainly of anthropogenic origin, being the result of high-temperature combustion processes.

As a consequence, the quantification of the NO_2 atmospheric level is important for understanding tropospheric pollution and provides information for air pollution monitoring, modelling and management. Different classes of

---

L. Aidaoui
Laboratoire de Développement en Mécanique et Matériaux,
University of Djelfa, Djelfa, Algeria
e-mail: aidaoui.ud@gmail.com

A. Maurizi (✉)
Institute of Atmospheric Sciences and Climate,
Consiglio Nazionale delle Ricerche,
Bologna, Italy
e-mail: a.maurizi@isac.cnr.it

A. Azzi
Laboratoire Aero-Hydrodynamique Navale, Faculté de Génie
Mécanique, Université des Sciences et de la Technologie
d'Oran (USTO), Oran, Algeria
e-mail: azzi.abbes@yahoo.fr

tools are available for this purpose. Ground-level measurement networks furnish detailed information on local surface concentration, while satellite instruments gives information at a large scale with global coverage (low earth orbit satellites—LEO), although limited to vertically integrated properties and also typically limited to one measurement per day (for LEO). Coupled models of atmospheric dynamics and chemistry are used in combination with measurements to obtain further information concerning the spatial and temporal distribution of pollutants (Lalitaporn et al. 2013), forecast pollutant concentrations (see, e.g. Kukkonen et al. 2012, for a recent review), and perform scenario studies (see, e.g. Colette et al. 2012).

In the last decade, an increasing number of studies have used both model simulations and satellite retrieved data for different purposes. In some cases, the two types of information are used in combination to give a more comprehensive picture and study specific features as in Curier et al. (2014), where tropospheric NO_2 concentration trends during 2005–2010 over Europe were derived from the ozone monitoring instrument (OMI) and LOTOS-EUROS model and compared to reported NOx emissions, or in Wang and Chen (2013), where a combination of model results and satellite data were used to derive surface concentrations of NO_2. However, a large part of the literature deals with model verification using satellite data as a reference, with the assessment of different simulation parameters on the model results. Huijnen et al. (2010) compared the ensemble median of the regional air quality (RAQ) models in the Global and regional Earth-system (Atmosphere) Monitoring using Satellite and in situ data (GE-MS) Project[1] with the tropospheric NO_2 vertical column density (VCD) from Dutch OMI NO_2 (DOMINO) 1.0.2, for the period July 2008–June 2009, over Europe. The spatial distribution was found to agree well with OMI observations, displaying a correlation of 0.8. A comparative study between OMI observations and CMAQ model simulations of tropospheric NO_2 VCD in East Asia (Han et al. 2011) was carried out over seasonal episodes in 2006, to evaluate the accuracy of the NOx emissions over the Korean peninsula, with correlation that ranges from 0.52 to 0.85, depending on the region and season considered. In Zyrichidou et al. (2013), the CAMx model is used to simulate NO_2 tropospheric VCD at high-resolution, which were evaluated and compared against both a previous study and OMI measurements over South-eastern Europe. The annual spatial correlation between OMI and the high-resolution model turned out to be 0.6, somewhat improved compared to a previous study (Zyrichidou et al. 2009). When using satellite data for model evaluation, it must be borne in mind that the satellite product itself is the result of an inversion process which, in turn, relies on model results. In fact, tropospheric vertical column density VCD is the result of direct observation of the bulk radiative effect of the observed quantity at given wavelengths, combined with a retrieval algorithm involving the use of a priori profiles derived from model results (Richter and Burrows 2002; Boersma et al. 2007). This is largely discussed when satellite products are compared (see, e.g. Zyrichidou et al. 2013) and taken into account when model performances are verified against satellite data (Zyrichidou et al. 2009; Yamaji et al. 2014). However, satellite products have become more reliable also as a consequence of the improvements in the inversion procedure. Since the mid-nineties, satellite remote sensing has been used to derive tropospheric NO_2 content at different atmospheric scales (Schaub et al. 2007; Hilboll et al. 2013; Huang et al. 2013), starting with GOME-1, then SCIAMACHY, OMI and GOME-2. Among these satellite instruments, OMI has a better spatial resolution (13×24 km^2 at nadir) than GOME (320×40 km^2) and SCIAMACHY (60×30 km^2), which makes it suitable for use in modelling studies where resolution can be relatively high.

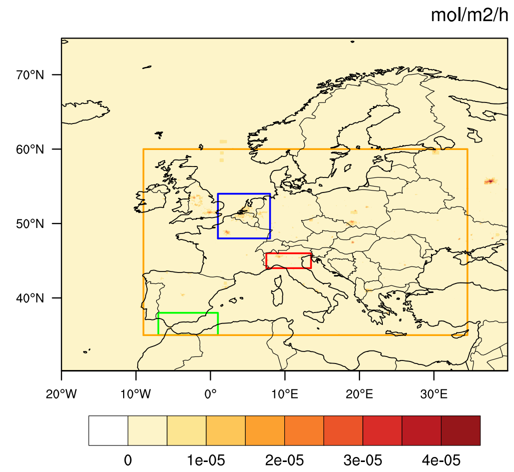


Fig. 1 Selected regions for analysis: Europe (*yellow*), Po Valley (*red*), BeNeLux (*blue*) and Gibraltar (*green*), overlapped to average NO_2 emission rates sample (unit: mol/m^2/h)

In the present study, the BOLCHEM model (Mircea et al. 2008; Maurizi et al. 2010) is employed to simulate NO_2 VCD with the aim of verifying the model's performances in highly polluted areas affected by different meteorological conditions, using different model resolutions. The DOMINO product v2.0[2] (Boersma et al. 2007) will be used for comparison. BOLCHEM started as an online coupled meteorology and composition model (Baklanov et al. 2014) in 2003 (Butenschoen et al. 2003), when this approach was relatively new. It was successfully used in the GEMS Project for operational forecasts of gas-phase pollutants as a member of the GEMS-RAQ ensemble, for over 1.5 years. In

---

[1] http://gems.ecmwf.int

[2] http://www.temis.nl/airpollution/no2.html

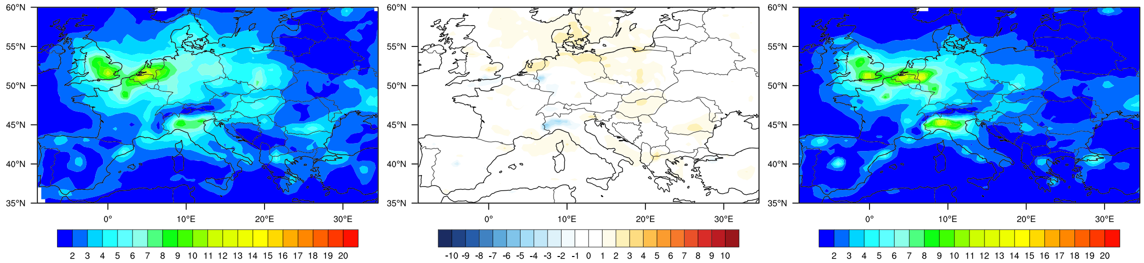


Fig. 2 Annual average of NO_2 VCD (unit: $\times 10^{15}$ molecules cm^{-2}) over Europe: model (*left*), model-satellite (*middle*) and satellite (*right*)

the same framework, it was compared indirectly, as part of the GEMS-RAQ ensemble, to satellite-retrieved NO_2 VCD (Huijnen et al. 2010) and, directly, with IASI tropospheric O3 columns (Zyryanov et al. 2012). Moreover, an upgraded version (2.0) was also verified in a 10-year experiment against surface data measurements as part of "megaCITY—Zoom for the Environment" (CITYZEN) project (Colette et al. 2011), showing good skills particularly for NO_2 surface concentration. This work focuses on two main aspects: beside the analysis of the general behavior over Europe, one of the main interest is on the ability of the model to capture the NO_2 content in the two most polluted European areas (PoValley and BeNeLux). In those highly populated regions, emissions of NOx change rapidly over very fine spatial scale (size of roads). This fact combined with the non-linear nature of NO_2 makes the model resolution an important parameter. The evaluation of the effects of resolution change is the second objective of the present work.

Model simulations were performed throughout 2007 (the last year of the 10-year CITYZEN experiment) over Europe and the two main hot-spots therein: the Po Valley and the BeNeLux area. In addition, the area of Gibraltar is also considered since, in the absence of other important pollutant sources, shipping emissions have a major role. Analyses will be discussed on annual and seasonal bases and the effect of resolution will be also addressed.

The manuscript is organised as follows: "Model description and experimental setup" gives an overview of BOLCHEM, including the experimental setup. "OMI data" section provides a short description of the OMI satellite instrument, the retrieval algorithm and the tropospheric NO_2 VCD characteristics. The spatial and temporal distribution of simulated and observed NO_2 VCD are discussed in "Results and discussion" section, with reference to the effects of model resolution. Finally, in "Conclusions" section, some conclusions are drawn.

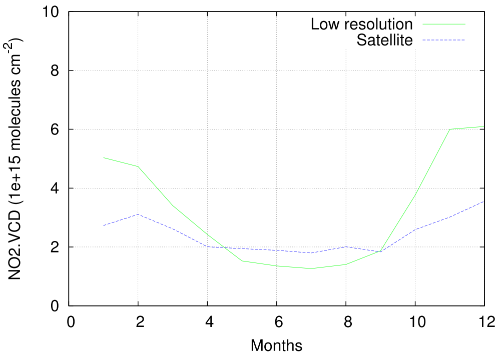


Fig. 3 Area average of NO_2 VCD over Europe (see Fig. 1) of the monthly mean tropospheric columns for 2007

Model description and experimental setup

The BOLCHEM model is an on-line coupled meteorology-composition model (Baklanov et al. 2014). Its meteorological component is the mesoscale meteorological BOlogna Limited Area Model (BOLAM)[3] (Buzzi et al. 1994, 2003), whose dynamics is based on hydrostatic primitive equations. The vertical grid uses a hybrid–terrain-following coordinate system, with variables distributed on a non-uniformly spaced staggered Lorenz (1960) grid. The horizontal discretisation uses geographical coordinates on an Arakawa C-grid. The time scheme is split-explicit, forward-backward for gravity modes. The weighted average flux (Billet and Toro 1997) advection scheme is implemented. Lateral boundary conditions are imposed using a relaxation scheme in order to minimise the wave energy reflection. As initial and lateral boundary conditions, use can be made of the data from the European Centre for Medium-range Weather Forecasts (ECMWF) or the global forecast system (NOAA-GFS.[4] Hybrid model level data are directly interpolated on the BOLAM grid. Transport (advection and diffusion) of tracers (both passive and reactive) is performed on-line at each meteorological time-step using the mass-conservative WAF scheme (Maurizi et al. 2013) for advection and a "physical" (second-order) horizontal-diffusion, with diffusion coefficient carefully estimated from experiments (Tampieri and Maurizi 2007) to account for unresolved motion. Vertical diffusion is performed using a one-dimensional diffusion equation with a diffusion coefficient

---

[3] http://www.isac.cnr.it/~dinamica/bolam/index.html

[4] http://www.ncdc.noaa.gov/data-access/model-data/model-datasets/global-forcast-system-gfs

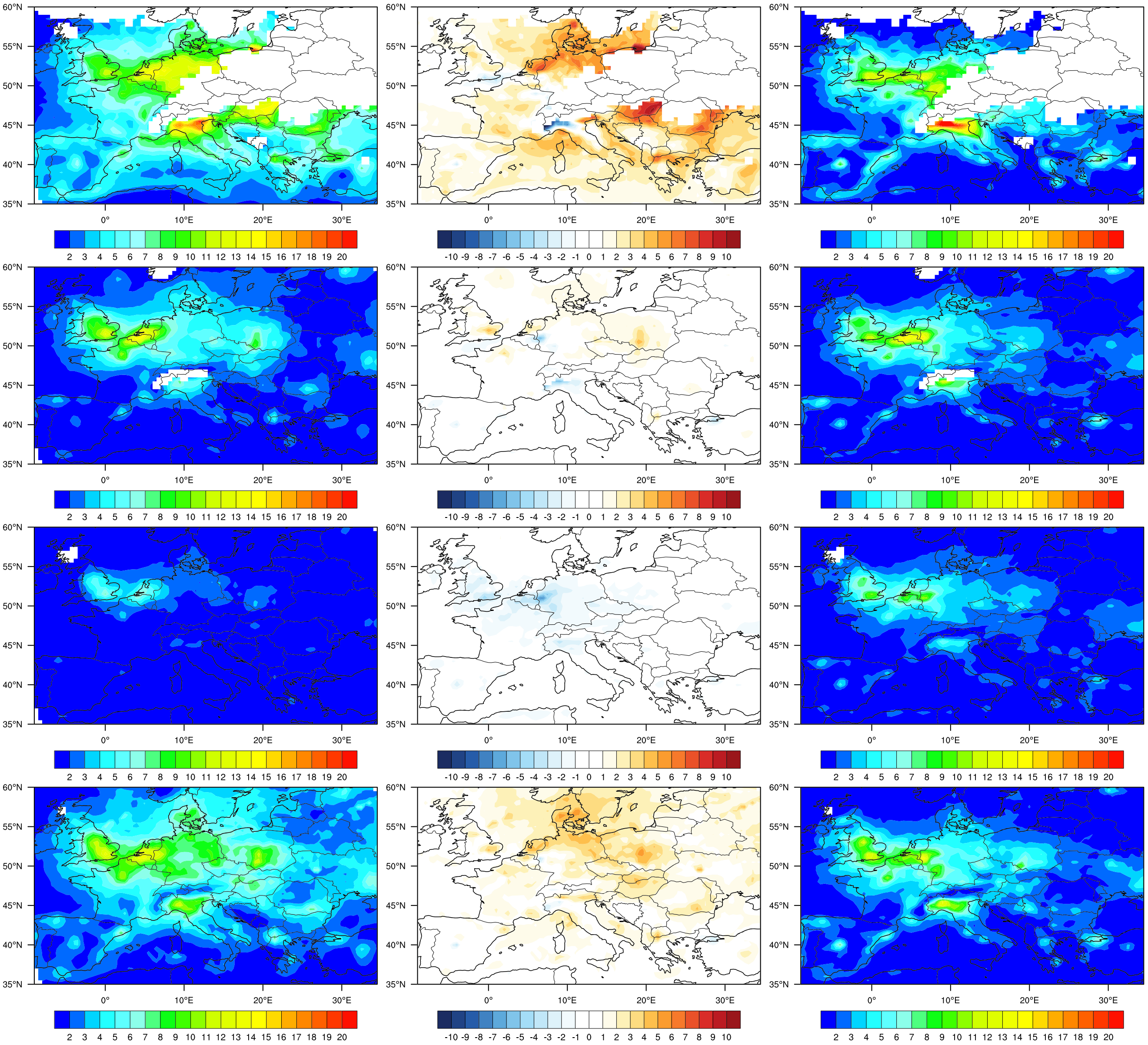


Fig. 4 Seasonal average of NO_2 VCD (Unit: $\times 10^{15}$ molecules cm^{-2}) over Europe: model (*left*), model-satellite (*middle*) and satellite (*right*). Seasons are winter, springer, summer and autumn from *top to bottom*, respectively. *White areas* for model and satellite maps represent missing values

estimated by means of an E-l turbulence closure scheme (Zampieri et al. 2005). Dry deposition is computed through a resistance-analogy scheme and is provided as boundary condition to the vertical diffusion equation. Furthermore, vertical redistribution of tracers due to moist convection is parameterised consistently with the Kain-Frisch scheme used in the meteorological component for moist convection. Transport of chemical species is performed in mass units, while gas chemistry is computed as mixing ratio. Physical/chemical processes are treated separately for gas phase, aerosol classes and generic tracers (e.g. radioactive species, Saharan dust, etc.). The gas phase is treated using the SAPRC90 mechanism modified to account for secondary organic aerosol precursors. The Aerosol component is modelled using AERO3. The model is included in the COST 728/732 model inventory,[5] where more technical details can be found.

Model runs were performed over Europe with a horizontal resolution of 50×50 km^2, and in the two hot-spots (Po Valley and BeNeLux, as defined for the CITYZEN project)

---

[5] http://www.mi.uni-hamburg.de/List-classification-detail-view.6156.0.html?&no_cache=1&mvid=2621441

Table 1 Annual and seasonal statistical indices computed over Europe and selected regions

		Low-res.			High-res.		
Region	Season	RMSE	MB	r	RMSE	MB	r
Europe	Annual	0.82	0.52	0.90	–	–	–
	Winter	2.97	2.23	0.76	–	–	–
	Spring	0.75	0.24	0.91	–	–	–
	Summer	0.82	−0.55	0.88	–	–	–
	Autumn	1.61	1.20	0.86	–	–	–
Po Valley	Annual	1.88	−0.46	0.74	1.39	0.11	0.91
	Winter	4.72	−0.11	0.46	5.13	2.58	0.69
	Spring	2.01	−1.23	0.83	1.66	−0.74	0.89
	Summer	1.51	−1.28	0.76	1.51	−1.30	0.81
	Autumn	2.03	1.03	0.81	2.10	1.13	0.86
BeNeLux	Annual	1.21	0.35	0.85	1.31	0.62	0.90
	Winter	2.81	1.80	0.61	2.69	1.44	0.66
	spring	1.40	0.40	0.88	1.71	0.95	0.91
	Summer	2.13	−1.81	0.75	1.75	−1.37	0.83
	Autumn	2.07	1.67	0.84	2.11	1.67	0.87
Gibraltar	Annual	0.50	0.31	0.79	–	–	–
	Winter	1.52	1.31	0.77	–	–	–
	spring	0.27	−0.06	0.79	–	–	–
	Summer	0.44	−0.32	0.60	–	–	–
	Autumn	0.67	0.45	0.76	–	–	–

Units for RMSE and MB is $\times 10^{15}$ molecules cm^{-2}

at 10×10 km^2, with 40 and 20 vertical sigma-hybrid levels for meteorology and chemistry, respectively. The top of chemistry domain is approximately at 500 h̃Pa. Equations are integrated on a rotated-pole coordinate system with a time-step of 400 s. Boundary conditions for the meteorology were supplied by ECMWF; for tracers, gas and aerosols, climatological boundary conditions were used for the European domain. For the two hot-spots, the boundary conditions were taken from the simulation of the European domain every hour. Emissions prepared for the CITYZEN project by INERIS[6] (Colette et al. 2011) were used. This emission dataset was prepared for the 10-year experiment using on official European Monitoring and Evaluation Programme (EMEP) data (with resolution of 50 × 50 km^2) and spatial distribution based on the GEMS-RAQ emissions (at 10 × 10-km^2 resolution). This emission dataset is affected by the typical uncertainty of similar datasets like, e.g. the one prepared with the same spatial resolution in the frame of the "Monitoring Atmospheric Composition and Climate (MACC)".[7]

OMI data

The ozone monitoring instrument (OMI) flies on the NASA/EOS-AURA satellite, providing the possibility of global measurements of the atmospheric NO_2 VCDs via measuring direct and backscattered sunlight in the ultraviolet-visible range from 270 to 500 nm (Levelt et al. 2006). The instrument was launched in July 2004 with a Sun-synchronous polar orbit crossing the equator at 13:30 local time. The OMI satellite observes the atmosphere with a spatial resolution of 13 km along track and 24 km across track in the nadir view, with a global coverage in 1 day.

The OMI tropospheric NO_2 level 2 data (Dutch OMI NO2, DOMINO v2.0) used in this study for Europe in 2007 are obtained from Tropospheric Emission Monitoring Internet Service (TEMIS) project.[8] According to Boersma et al. (2007) and Boersma et al. (2011), the NO_2 DOMINO retrieval algorithm is based on three main steps: (i) obtain the NO_2 slant column density from OMI reflectance spectra, using differential optical absorption spectroscopy (DOAS); (ii) estimate and separate the stratospheric and tropospheric contributions to the slant column; and (iii) convert the remaining tropospheric slant column to a vertical column

---

[6] http://www.ineris.fr/

[7] https://www.gmes-atmosphere.eu/

[8] http://www.temis.nl

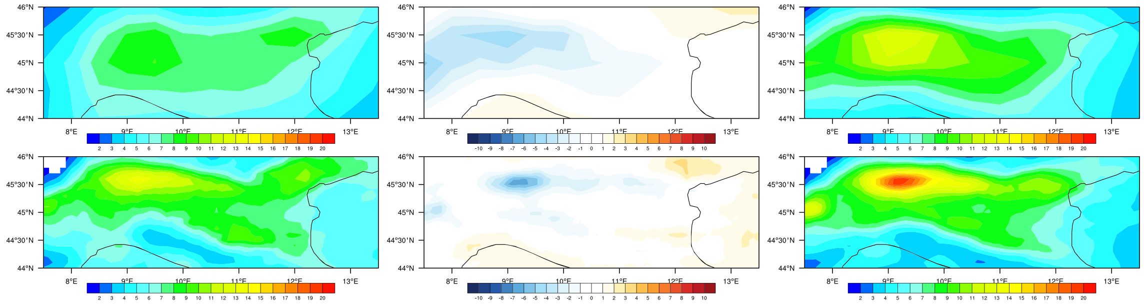


Fig. 5 Annual average of NO_2 VCD (unit: $\times 10^{15}$ molecules cm^{-2}) over Po Valley: model (*left*), model-satellite (*middle*) and satellite (*right*), for low-resolution (*top*) and high-resolution (*bottom*)

using the tropospheric air mass factor (AMF). Moreover, only satellite measurements with cloud fractions less than 50 % are included in the satellite dataset (van der A et al. 2008).

Further information on the DOMINO v2.0 retrieval algorithm is available in Boersma et al. (2011) and the latest updates can be found in the DOMINO product specification document.[9]

Results and discussion

Comparison of model and satellite data were performed for the whole of Europe with focus on selected regions (Fig. 1) at two different resolutions: 50 × 50 and 10 × 10 km^2. The analysis was performed on annual, seasonal and monthly basis. For the seasonal analysis, conventional seasons were used: winter (January, February, December), spring (March, April, May), summer (June, July, August) and autumn (September, October, November). Notice that winter is based on three months of the same year.

Satellite level 2 *swath* files were regridded on daily basis using the WHIPS[10] tool, onto common analysis grids with the same model resolution: 50 × 50 km^2 in the Europe and Gibraltar cases and 10 × 10 km^2 for the Po Valley and BeNeLux. In the regridding procedure, data with cloud cover larger than 0.2, surface albedo larger than 0.3 and solar zenith angle larger than 85° are discarded. This filter produces a mask of valid measurements that is applied also to model results to make the satellite and model datasets fully consistent.

The model output was sampled at the same satellite overpass time over Europe (13:30 UTC). The modelled VCD was computed using the averaging kernel (AK) corrected by the ratio between the total and the tropospheric AMF (Eskes and Boersma 2003). Because model simulation reach a height of about 500 hPa, the upper NO_2 content was extrapolated linearly from the upper model value to zero at the tropopause (where O3 exceeds 150 ppb which, above Europe, corresponds to about 200 hPa) (Huijnen et al. 2010). The linear extrapolation to zero is well-supported by the climatological fields used in CITYZEN (Colette et al. 2011) and the analysis about the application of the AK found in Huijnen et al. (2010).

Low resolution: Europe and selected regions

Results of the analysis of annual and seasonal tropospheric NO_2 VCD as simulated by the BOLCHEM model and observed by OMI over Europe are reported in Figs. 2, 3 and 4 and Table 1.

In this section, the main features are analysed for the whole Europe and selected regions: Po Valley, BeNeLux and Gibraltar (Fig. 1), at coarse resolution. Yearly and seasonal analyses are also discussed. Figure 2 shows the annual spatial distribution of NO_2 VCD over Europe of the DOMINO product (right) and the model output (left), along with the difference between them (middle).

At a first glance, a good agreement between model and satellite can be noticed (Fig. 2). Similar features are displayed by predicted and simulated VCD over highly polluted regions (the Po Valley and BeNeLux hot-spots and the largest European cities; Paris, London, Madrid, Barcelona, Rome, etc.). Generally speaking, the model presents a background higher and smoother than the satellite, mainly in winter and autumn. The model underestimates VCD over some large cities (Cologne, Istanbul, Madrid, Milan, Turin), while displaying a weak overestimation over others (Athens, Barcelona, London, Marseilles, Paris, Rome) and a general overestimation over the Netherlands. Looking at the two hot-spots, it can be seen that VCD in the Po Valley is underestimated by the model, while the behavior for BeNeLux is less uniform, displaying both over- and under-estimations.

Model underestimation observed in the Po Valley (Fig. 5 upper panel) can be related to the complex Alpine topography and its impact on the data retrieval

---

[9] http://www.temis.nl/docs/OMI_NO2_HE5_2.0_2011.pdf

[10] http://www.sage.wisc.edu/download/WHIPS/WHIPS.html

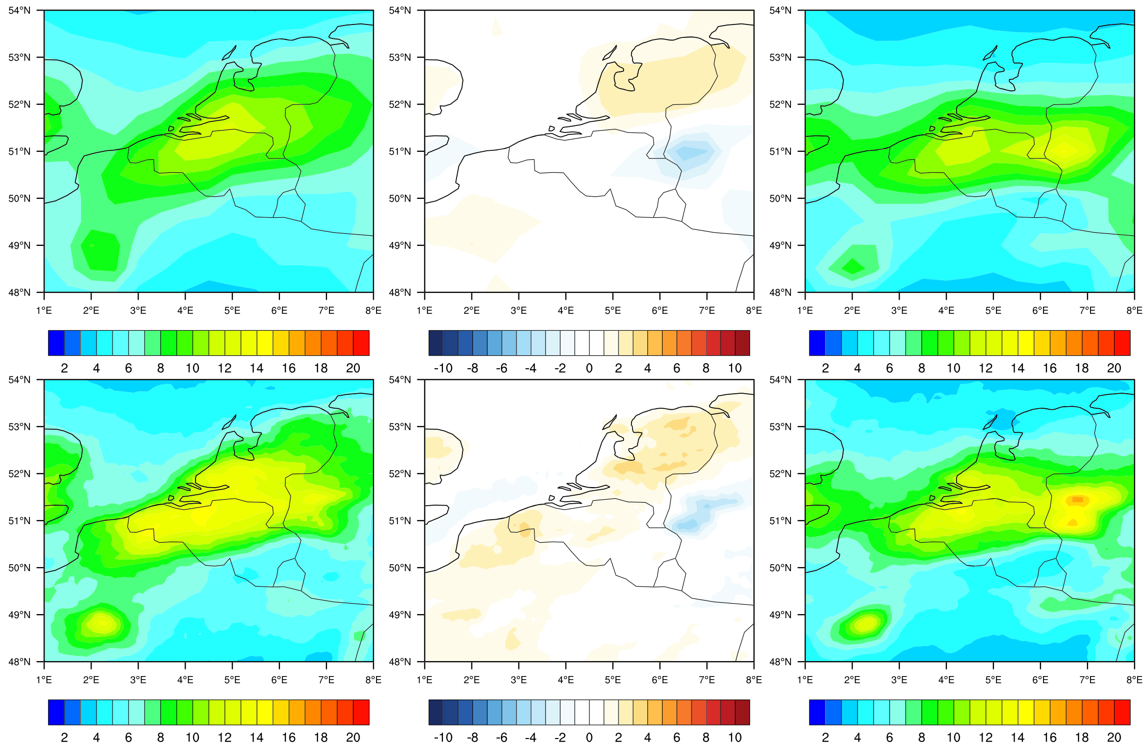


Fig. 6 Annual average of NO_2 VCD (unit: $\times 10^{15}$ molecules cm^{-2}) over BeNeLux: model (*left*), model-satellite (*middle*) and satellite (*right*), for low-resolution (*top*) and high-resolution (*bottom*)

(Schaub et al. 2007; Zhou et al. 2009), in addition to inaccuracies in the emissions inventory. In contrast to this seasonal change observed over Europe, the underestimation in the Po Valley is relatively uniform over time.

In the BeNeLux area (Fig. 6 upper panel), a mixture of slight over- and under-estimation of VCD is observed. The satellite map presents two maxima while the model only has one, missing the maximum observed over the Cologne area.

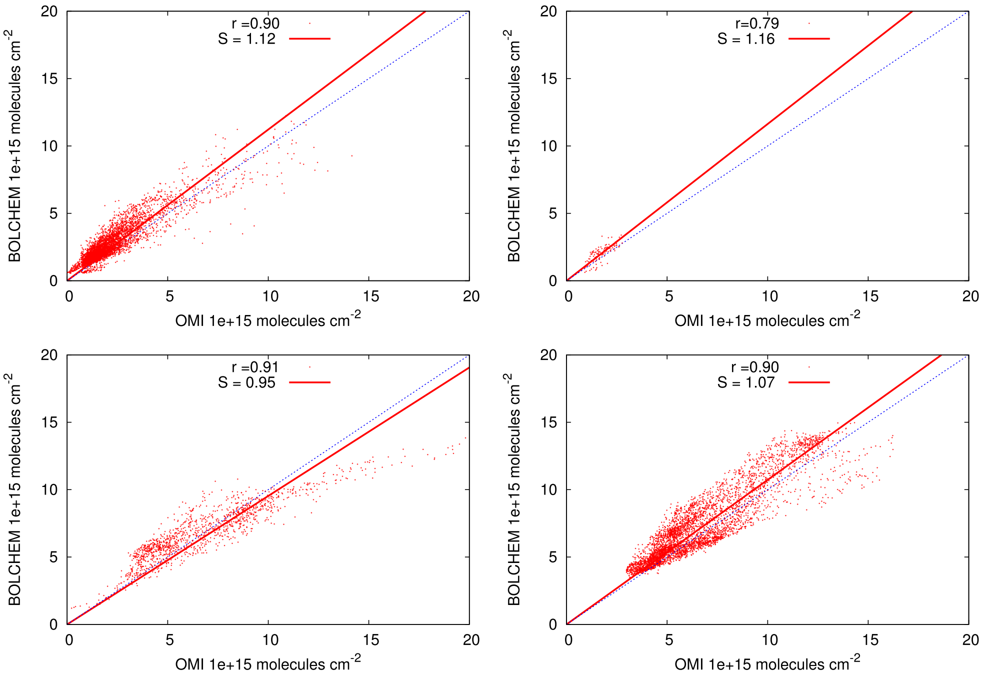


Fig. 7 Scatter plots between BOLCHEM and OMI NO_2 columns: **a** Europe, **b** Gibraltar at low-resolution, **c** Po Valley and **d** BeNeLux at high-resolution. r is the correlation coefficient and S is the slope

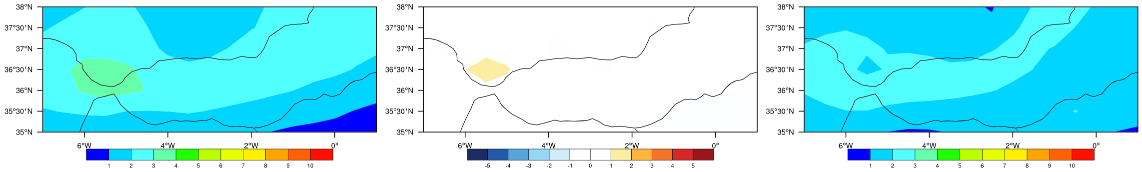


Fig. 8 Annual average of NO_2 VCD (unit: $\times 10^{15}$ molecules cm^{-2}) over Gibraltar: *left* (model), *centre* (difference: model-satellite) and *right* (satellite)

This is likely to be caused by inaccuracies in the emission database. The effect of finer resolution will be discussed in "High resolution over Po Valley" and "High resolution over BeNeLux" sections.

Looking at the south-west part of the domain, the ship tracks are more visible in the model map, especially for the Gibraltar area, while the satellite map shows some signs of the North African big cities and industrial areas (e.g. Algiers, Oran, S'kikda, Tunis) that are not visible in the model map, clearly reflecting the lack of emissions in the southern part of the simulated domain (North Africa).

Annual and seasonal root mean square error (RMSE), mean bias (MB) and correlation (r) were computed for Europe and specific regions (see Table 1). The spatial correlation of the annual maps between OMI DOMINO v2.0 and BOLCHEM is 0.90. The value of the above correlation coefficient is high compared to typical values found in the literature.

Seasonal average maps of VCD are presented and compared in Fig. 4, and a statistical analysis is reported in Table 1. Figure 4 is organised in the same way as Fig. 2: model results in the left panel, satellite observations in the right panel and differences in the middle.

Figure 4 shows a good agreement between model and satellite data with hot-spots and urban polluted patterns similarly distributed in spring and summer, with spatial correlation coefficients (Table 1) of 0.91 and 0.88, respectively, and small RMSE and MB.

In autumn and winter, there is a general overestimation more pronounced near areas were missing values are present, possibly highlighting some efficiency problem in the filtering procedure. In particular, the large areas of missing values observed in north-east Europe result from the combination of cloudiness and high surface albedo due to snow cover. Selecting thresholds higher than the "standard ones" would enlarge the missing data areas, thus removing the large overestimation in winter over, e.g. Hungary. Underestimation is observed in some spots like Madrid, Frankfurt and the English Channel, with a strong maximum in the Po Valley in which, however, high overestimation in winter is observed in pixels adjacent to missing data areas and can be partly remove with more strict thresholds. Despite this, the spatial correlation over Europe is still high (0.86) in autumn and acceptable in winter (0.76).

Looking at specific regions, it is observed that (Table 1) the correlation is moderately higher for BeNeLux than for the Po Valley and Gibraltar. The correlation is good in the Po Valley, except in winter when the area is frequently covered by clouds. Gibraltar presents also good correlation, except in summer, in contrast to the lowest value observed in winter in the other two regions. Concerning the MB, the annual values, which differ for each area, are unevenly distributed over time, with typically small negative values in spring/summer and high positive values in winter and autumn, except for the Po Valley where even winter values are very small, due to a compensation between the underestimation in the west and the overestimation in the east.

Figure 7 shows the scatter-plots between the BOLCHEM simulated and the OMI retrieved tropospheric NO_2 VCD over Europe, Gibraltar for low-resolution (50×50 km^2) and the Po Valley, BeNeLux for high-resolution (10×10 km^2). For Europe, most of the data is above the 1:1 line, which reflects the already noted annual overestimation, with the

Fig. 9 Area average of NO_2 VCD over Gibraltar (see Fig. 1) of the monthly mean tropospheric columns for 2007 (*left panel*). In the *right panel*, relative differences are shown

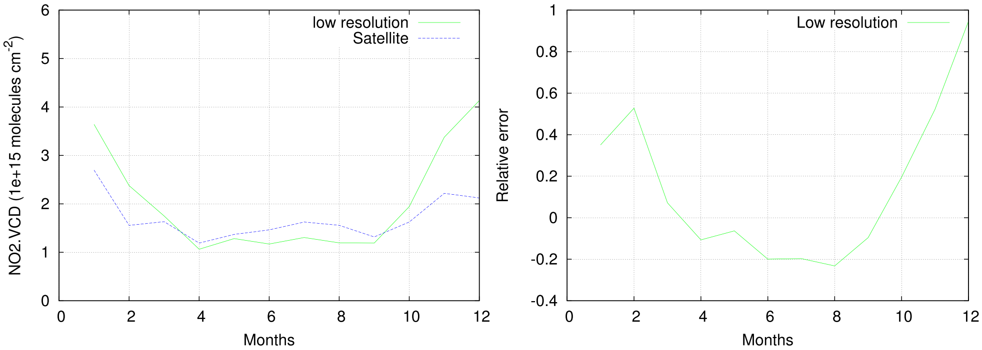
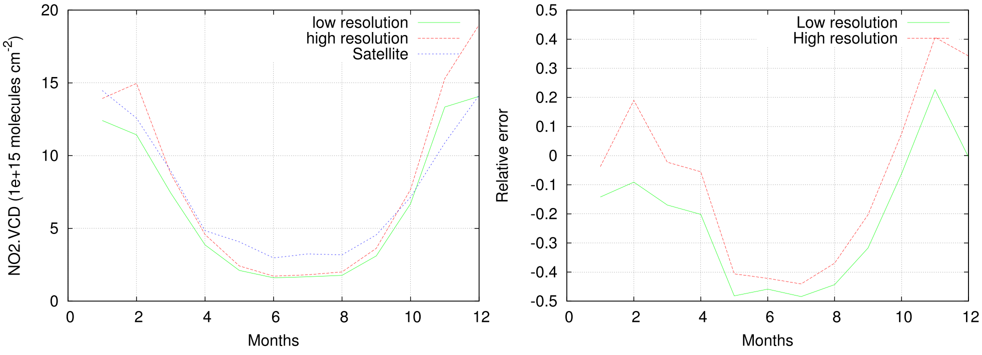


Fig. 10 Area average of NO_2 VCD over Po Valley (see Fig. 1) of the monthly mean tropospheric columns for 2007 with different model resolutions (*left panel*). In the *right panel*, relative differences are shown



subset of data for north-eastern Europe clearly identifiable, and well above the regression line. Clear overestimation is also apparent in the BeNeLux and Gibraltar scatter-plots. In the Po Valley, the scatter-plot displays an overestimation for small values and an underestimation for the highest end of the VCD range, which corresponds to the model underestimation over the highly polluted Milan area observed in Fig. 5.

Despite Gibraltar was not the focus of the CITYZEN project, model simulation cover also that region which is highly affected by shipping emission. Although the domain is located near the boundary and is probably influenced by the boundary conditions, which in the present simulations are climatological, the area is mostly affected by shipping emissions which are present in the emission dataset used for the model simulations and, therefore, the analysis of results can provide useful information.

Figure 8 displays NO_2 VCDs for the Gibraltar region, which are lower than in the other selected regions, this area not being affected by very large emissions. VCD values range from 100 to 400 $\times 10^{15}$ molecules cm^{-2} during the whole of 2007 for both model and satellite. The lack of emissions in North Africa, already observed when analysing results for Europe, do not seem to play a major role. In general, the most influential sources in the area seem to be the southern Spain cities and shipping emissions. The model predicts higher values of VCD than the satellite, in agreement with previous findings where a strong model overestimation is observed over the sea in correspondence to shipping routes (Im et al. 2014).

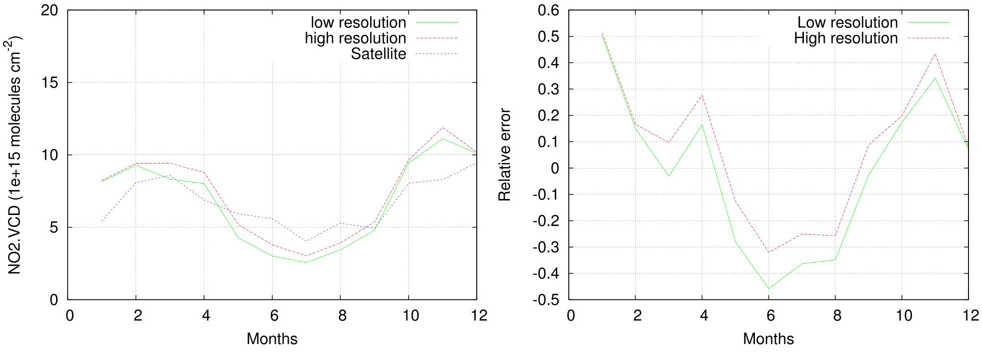
Looking at the annual cycle (Fig. 9), during spring and summer, there is a small difference between the spatial average of OMI measured and model simulated NO_2 VCDs, ranging from 5 to 8 % in spring and is 18 % in summer. In winter and autumn, a model overestimation is observed.

A more detailed analysis is found in Table 1. The statistical analysis displays a small magnitude of the Mean Bias in spring and summer with -0.06 and -0.32 $\times 10^{15}$ molecules cm^{-2}, respectively. It is worth noting that an unusual seasonal spatial correlation (high in winter and small in summer) appears in the seasonal statistical analysis (Table 1) in contrast to what happens for the other selected regions. It must be borne in mind, however, that this region is not only less affected by large emissions but also presents low cloudiness.

High resolution over Po Valley

In this section, we present a comparison of DOMINO v2.0 with coarse- and high-resolution simulated tropospheric NO_2 VCDs from the BOLCHEM model over the Po Valley

Fig. 11 Area average of NO_2 VCD over BeNeLux (see Fig. 1) of the monthly mean tropospheric columns for year 2007 with different model resolutions (*left panel*). In the *right panel*, relative differences are shown



hot-spot. The discussion is based on the results reported in Figs. 5 and 10 and Table 1.

Figure 5 displays a generally good agreement between model and satellite for the annual average, especially for the high-resolution simulation. The same spatial features, which changes significantly with resolution, are discerned in both the model and satellite such as the high NO_2 amounts for Milan and low amounts over the Alps and Appenines. Model overestimation in Liguria (west of Genoa) and over the Ligurian Sea is observed. Concerning Liguria, the overestimation is possibly caused by the poor representation of the Maritime Alps barrier between the Po Valley and Ligurian Sea in the meteorological model, allowing for some spurious transport from the more polluted Po Valley area towards the sea. Underestimation is generally reduced except over Milan. The annual spatial correlation is $r = 0.91$ (Table 1), a clear improvement with respect to the low-resolution correlation ($r = 0.74$, Table 1).

Figure 10a presents the time series of the monthly mean VCD from OMI satellite measurements and model-predicted values averaged over the Po Valley region. The model is quite successful in capturing the VCD annual cycle, with small absolute differences, except in winter. Looking at the relative errors, the high-resolution simulated VCD values are larger than the OMI ones by 0 to 35 % during winter time. In summer, there is an underestimation of more than 40 %, enhanced by the low absolute values of VCD. A different picture emerges at coarse-resolution, which displays a better agreement in winter and autumn, with relative errors ranging from 0 to −0.15 in winter and −0.43 to −0.48 in summer. The bulk effect of increasing resolution is to increase the simulated NO_2 VCD by 5 to 30 % for the whole year. This seems to be effective in reducing the error mainly in summer, when the photochemical reactions are dominant.

A good correlation is found for the Po Valley for all seasons in the high-resolution experiment (Table 1), higher than those found for the low-resolution experiment (Table 1) with a larger improvement in winter in which the lowest value is found for both resolutions. The mean bias shows variations with season for high-resolution, with the greatest value in winter (2.58×10^{15} molecules cm^{-2}). This highlights the better agreement between model and satellite observations in cloud-free conditions.

The seasonal statistical analysis in Table 1 demonstrates the positive effect of the high-resolution model in spring and summer and the negative effect during winter and autumn. Table 1 displays high RMSE in winter at both resolutions. The magnitude of the mean bias decreases with high-resolution from 1.23 to 0.74×10^{15} molecules cm^{-2} in spring, whereas the same parameter increases from 0.11, 1.03 to 2.58 and 1.13×10^{15} molecules cm^{-2} for winter and autumn, respectively.

High resolution over BeNeLux

As for the Po Valley, the BeNeLux model results at two different spatial resolutions are compared with satellite data. In Fig. 6, which shows a close-up of the BeNeLux region, the spatial distribution (maxima and minima locations) of NO_2 VCDs is not very well captured by the model at low resolutions because model only presents a maximum of VCD while satellites displays two. This picture is slightly improved by the high-resolution that displays a secondary maximum in correspondence to the Rhine-Ruhr area, the main center of European steel production and the big inland port of Duisburg. However, VCD in this area is slightly underestimated at both resolution with a more pronounced effect at low-resolution (Fig. 6b, e). On the other end, high-resolution simulation displays larger overestimation in areas not affected by large emissions. Moreover, as a general comment, the model always displays a smoothly distributed VCD, unlike the satellite VCD, which is more concentrated in areas of high emissions.

In contrast to the results for the sea in other areas, a large NO_2 VCD is observed by both model and satellite between the UK and Belgium, and more generally (see Figs. 2 and 4) in the whole English Channel and south of the North Sea. This is possibly because, in addition to shipping emissions, a large contribution to the high level of pollution comes from the highly polluted adjacent land areas.

The spatial correlation is improved in high-resolution simulation (Table 1) uniformly over the seasons. The statistical analysis shows that the high-resolution model has positive effects during summer time in agreement with Yamaji et al. (2014). This is more evident in this region than in the Po Valley. According to the relative error plot (Fig. 11b), the high-resolution simulation reduces the error between simulated and measured VCDs by 29 % in summer.

As shown in Table 1, MB displays strong seasonal variations. The RMSE varies slightly, displaying higher values in winter and autumn and lower in spring and summer with only slight variations between high and low resolution.

Conclusions

Simulations of atmospheric pollutants for the year 2007 over Europe with a focus on hot-spots at higher resolution were analysed in order to verify the ability of the BOLCHEM model to reproduce the NO_2 vertical column density retrieved from OMI (DOMINO v2.0 product from TEMIS). The analysis was performed on an annual, seasonal and monthly basis.

The model turned out to perform well, with statistical parameters in line with those found in the literature for other comparison experiments (e.g. Han et al. 2011; Zyrichidou et al. 2013, Im et al. 2014) with particularly high values of the correlation coefficient. Analysis on a seasonal basis confirms the good BOLCHEM performances, while revealing a difference between the “warm” and “cold” seasons: in spring and summer, performances are usually better than in winter. However, the statistical significance of winter satellite data is reduced by the low number of observations caused by the presence of clouds and also by increased surface albedo due to snow cover.

Performances in the low polluted region of Gibraltar, where one of the main sources is shipping emissions, is relatively good despite the missing emissions from North Africa and also possibly by the boundary conditions can have a negative influence. At variance with the other analysed regions, the spatial correlation is higher in winter and lower in summer when photochemistry is active.

The analysis of high-resolution simulations over the two main European hot-spots reveals that high-resolution is more effective in the Po Valley, where it increases the spatial correlation more than in the BeNeLux. However, the effect on the other scores is less clear. The increase of correlation is an important signature of the impact of non-linearities in photochemistry. The observed differences between the Po Valley and BeNeLux in response to increasing resolution can be explained by the large differences in meteorology. In BeNeLux, where wind is typically stronger than in the Po Valley and causes more mixing which, in turn, is expected to make the concentrations less dependent on local emissions, high resolution does not seem to increase the model prediction. This is confirmed by the differences observed in the seasonal analysis, which shows a positive impact in “warm” seasons, where photochemistry is more effective, and negative in “cold” ones.

An extended analysis, also involving surface data and ground-based remote sensing with profiling capabilities, would be an interesting step towards understanding both satellite data reliability and model performances with regard to the effect of vertical distribution, which is known to be an important issue for both models and satellites.

Acknowledgments We acknowledge the free use of tropospheric NO_2 column data from the OMI sensor from `www.temis.nl`. The software used for the production of this article (model development, model run, data analysis, graphics, typesetting) is Free Software. The authors would like to thank the whole free software community, the Free Software Foundation[11] and, in particular, the Debian Project.[12] This is part of the PhD research project of Lakhdar Aidaoui supported by the Ministry of High Education and Scientific Research of Algeria. The authors would like to thank Dr. Bianca Maria Dinelli and Dr. Enrico Arnone for helpful discussion on the analysis of satellite data. The research leading to these results has received funding from the European Unions Seventh Framework Programme (FP7/2007-2013) under grant agreement no. 212095 (CityZen).

---

[11] http://www.fsf.org

[12] http://www.debian.org

References

Ashmore MR (2005) Assessing the future global impacts of ozone on vegetation. Plant Cell and Environment 28(8):949–964. doi:10.1111/j.1365-3040.2005.01341.x

Baklanov A, Schlunzen K, Suppan P, Baldasano J, Brunner D, Aksoyoglu S, Carmichael G, Douros J, Flemming J, Forkel R, Galmarini S, Gauss M, Grell G, Hirtl M, Joffre S, Jorba O, Kaas E, Kaasik M, Kallos G, Kong X, Korsholm U, Kurganskiy A, Kushta J, Lohmann U, Mahura A, Manders-Groot A, Maurizi A, Moussiopoulos N, Rao ST, Savage N, Seigneur C, Sokhi RS, Solazzo E, Solomos S, Sørensen B, Tsegas G, Vignati E, Vogel B, Zhang Y (2014) Online coupled regional meteorology chemistry models in Europe: current status and prospects. Atmos Chem Phys 14(1):317–398. doi:10.5194/acp-14-317-2014

Billet SJ, Toro EF (1997) On WAF-Type schemes for multidimensional hyperbolic conservation laws. J Comput Phys 130:1–24

Boersma KF, Eskes HJ, Veefkind JP, Brinksma EJ, van der ARJ, Sneep M, van den Oord GHJ, Levelt PF, Stammes P, Gleason JF, Bucsela EJ (2007) Near-real time retrieval of tropospheric NO2 from OMI. Atmos Chem Phys 7:2103–2118

Boersma KF, Eskes HJ, Dirksen RJ, van der ARJ, Veefkind JP, Stammes P, Huijnen V, Kleipool QL, Sneep M, Claas J, Leitao J, Richter A, Zhou Y, Brunner D (2011) An improved tropospheric NO2 column retrieval algorithm for the Ozone Monitoring Instrument. Atmos Meas Tech 4:1905–1928

Butenschoen M, Maurizi A, Tampieri F (2003) Bolchem 0.9: A 3d prediction model of the meteorology and the chemical composition of the atmosphere. Tech. rep. CNR-ISAC, Italy

Buzzi A, Fantini M, Malguzzi P, Nerozzi P (1994) Validation of a Limited Area Model in cases of Mediterranean cyclogenesis: surface fields and precipitation scores. Meteorol Atmos Phys 53:137–153

Buzzi A, D’Isidoro M, Davolio S (2003) A case-study of an orographic cyclone south of the Alps during the MAP SOP. Q J Roy Meteor Soc 129–591:1795–1818

Chen R, Samoli E, Wong CM, Huang W, Wang Z, Chen B, Kan H (2012) Associations between short-term exposure to nitrogen dioxide and mortality in 17 Chinese cities: The China Air Pollution and Health Effects Study (CAPES). Environ Int 45:32–38

Colette A, Granier C, Hodnebrog Ø, Jakobs H, Maurizi A, Nyiri A, Bessagnet B, D’Angiola A, D’Isidoro M, Gauss M, Meleux F, Memmesheimer M, Mieville A, Rouil L, Russo F, Solberg S, Stordal F, Tampieri F (2011) Air quality trends in Europe over the past decade: a first multi-model assessment. Atmos Chem Phys 11:11,657–11,678. doi:10.5194/acpd-11-19029-2011

Colette A, Granier C, Hodnebrog Ø, Jakobs H, Maurizi A, Nyiri A, Rao S, Amann M, Bessagnet B, DAngiola A, Gauss M, Heyes C, Klimont Z, Meleux F, Memmesheimer M, Mieville A, Rouil L, Russo F, Schucht S, Simpson D, Stordal F, Tampieri F, Vrac M (2012) Future air quality in Europe: a multi-model assessment of projected exposure to ozone. Atmos Chem Phys 12(21):10,613–10,630. doi:10.5194/acp-12-10613-2012

Curier RL, Kranenburg R, Segers AJ, Timmermans RMA, Schaap M (2014) Synergistic use of OMI NO2 tropospheric columns and LOTOSEUROS to evaluate the NOx emission trends across Europe. Remote Sens Environ 149:58–69

Eskes HJ, Boersma KF (2003) Averaging kernels for doas total-column satellite retrievals. Atmos Chem Phys 3:1285–1291. doi:10.5194/acp-3-1285-2003, 2003

Fuhrer J, Skarby L, Ashmore MR (1997) Critical levels for ozone effects on vegetation in Europe. Environ Pollut 97(1–2):91–106. doi:10.1016/S0269-7491(97)00067-5

Han KM, Lee CK, Lee J, Kim J, Song CH (2011) A comparison study between model-predicted and OMI-retrieved tropospheric NO2 columns over the Korean peninsula. Atmos Environ 45:2962–2971

Hilboll A, Richter A, Burrows JP (2013) Long-term changes of tropospheric NO2 over megacities derived from multiple satellite instruments. Atmos Chem Phys 13:4145–4169

Huang J, Zhou C, Lee X, Bao Y, Zhao X, Fung J, Richter A, Liu X, Zheng Y (2013) The effects of rapid urbanization on the levels in tropospheric nitrogen dioxide and ozone over East China. Atmos Environ 77:558–567

Huijnen V, Eskes HJ, Poupkou A, Elbern H, Boersma KF, Foret G, Sofiev M, Valdebenito A, Flemming J, Stein O, Gross A, Robertson L, D'Isidoro M, Kioutsioukis I, Friese E, Amstrup B, Bergstrom R, Strunk A, Vira J, Zyryanov D, Maurizi A, Melas D, Peuch VH, Zerefos C (2010) Comparison of OMI NO2 tropospheric columns with an ensemble of global and European regional air quality models. Atmos Chem Phys 10:3273–3296

Im U, Daskalakis N, Markakis K, Vrekoussis M, Hjorth J, Myriokefalitakis S, Gerasopoulos E, Kouvarakis G, Richter A, Burrows J, Pozzoli L, Unal A, Kindap T, Kanakidou M (2014) Simulated air quality and pollutant budgets over Europe in 2008. Sci Total Environ 470–471:270–281

Kukkonen J, Olsson T, Schultz DM, Baklanov A, Klein T, Miranda AI, Monteiro A, Hirtl M, Tarvainen V, Boy M, Peuch VH, Poupkou A, Kioutsioukis I, Finardi S, Sofiev M, Sokhi R, Lehtinen KEJ, Karatzas K, San Josė R, Astitha M, Kallos G, Schaap M, Reimer E, Jakobs H, Eben K (2012) A review of operational, regional-scale, chemical weather forecasting models in Europe. Atmos Chem Phys 12(1):1–87. doi:10.5194/acp-12-1-2012

Lalitaporn P, Kurata G, Matsuoka Y, Thongboonchoo N, Surapipith V (2013) Long-term analysis of no 2, co, and aod seasonal variability using satellite observations over asia and intercomparison with emission inventories and model. Air Qual Atmos Health 6:655–672

Latza U, Gerdes S, Baur X (2009) Effects of nitrogen dioxide on human health: systematic review of experimental and epidemiological studies conducted between 2002 and 2006. Int J Hyg Environ Health 212:271–287

Levelt PF, van denOord GHJ, Dobber MR, Malkki A, Visser H, de Vries J, Stammes P, Lundell JOV, Saari H (2006) The ozone monitoring instrument. IEEE Trans Geosci Remote Sens 44:1093–1100

Lorenz EN (1960) Energy and numerical weather prediction. Tellus 12:364–373

Maurizi A, D'Isidoro M, Mircea M (2010) BOLCHEM, an integrated system for atmospheric dynamics and composition. In: Baklanov A, Mahura A, Sokhi R (eds) Integrated systems of mesometeorological and chemical transport models, vol 8. Springer, p 186

Maurizi A, Russo F, Tampieri F (2013) Local vs. external contribution to the budget of pollutants in the Po Valley (Italy) hot spot. Sci Total Environ 458–460:459–465

Mircea M, D'Isidoro M, Maurizi A, Vitali L, Monforti F, Zanini G, Tampieri F (2008) A comprehensive performance evaluation of the air quality model BOLCHEM to reproduce the ozone concentrations over Italy. Atmos Environ 42(6):1169–1185

Monks PS, Granier C, Fuzzi S, Stohl A, Williams ML, Akimoto H, Amann M, Baklanov A, Baltensperger U, Bey I, Blake N, Blake RS, Carslaw K, Cooper OR, Dentener F, Fowler D, Fragkou E, Frost GJ, Generoso S, Ginoux P, Grewe V, Guenther A, Hansson, HC, Henne S, Hjorth J, Hofzumahaus A, Huntrieser H, Isaksen ISA, Jenkin ME, Kaiser J, Kanakidou M, Klimont Z, Kulmala M, Laj P, Lawrence MG, Lee JD, Liousse C, Maione M, McFiggans G, Metzger A, Mieville A, Moussiopoulos N, Orlando JJ, CD ODowd, Palmer PI, Parrish DD, Petzold A, Platt U, Poschl U, Prevo ASH, Reeves CE, Reimann S, Rudich Y, Sellegri K, Steinbrecher R, Simpson D, ten Brink H, Theloke J, van der Werf GR, Vautard R, Vestreng V, Vlachokostas C, von Glasow R (2009) Atmospheric composition change global and regional air quality. Atmos Environ 43:5268–5350

Richter A, Burrows JP (2002) Tropospheric NO2 for GOME measurements. Adv Space Res 29(11):1673–1683

Richter A, Burrows JP, Nuß H, Granier C, Niemeier U (2005) Increase in tropospheric nitrogen dioxide over China observed from space. Nature 437:129–132

Schaub D, Brunner D, Boersma KF, Keller J, Folini D, Buchmann B, Berresheim H, Staehelin J (2007) SCIAMACHY tropospheric NO2 over Switzerland: estimates of NOx lifetimes and impact of the complex Alpine topography on the retrieval. Atmos Chem Phys 7:5971–5987

Solomon S, Portmann RW, Sanders RW, Daniel JS (1999) On the role of nitrogen dioxide in the absorption of solar radiation. J Geophys Res 104:12,047–12,058

Tampieri F, Maurizi A (2007) Evaluation of the dispersion coefficient for numerical simulations of tropospheric transport. Nuovo Cimento C 30:395–406

van der A RJ, Eskes HJ, Boersma KF, van Noije TPC, van Roozendael M, De Smedt I, Peters DHMU, Meijer EW (2008) Trends, seasonal variability and dominant NOx source derived from a ten year record of NO2 measured from space. J Geophys Res 113:D04,302. doi:10.1029/2007JD009021

Wang B, Chen Z (2013) An intercomparison of satellite-derived ground-level NO2 concentrations with GMSMB modeling results and in-situ measurements - A North American study. Environ Pollut 181:172–181

Yamaji K, Ikeda K, Irie H, Kurokawa J, Ohara T (2014) Influence of model grid resolution on no 2 vertical column densities over east asia. J Air Waste Manag Assoc 64:436–444

Zampieri M, Malguzzi P, Buzzi A (2005) Sensitivity of quantitative precipitation forecasts to boundary layer parameterization: a flash flood case study in the western mediterranean. Nat Hazards Earth Syst Sci 5:603–612

Zhou Y, Brunner D, Boersma KF, Dirksen R, Wang P (2009) An improved tropospheric NO2 retrieval for OMI observations in the vicinity of mountainous terrain. Atmos Meas Tech 2:401–416

Zyrichidou I, Koukouli ME, Balis D, Katragkou E, Melas D, Poupkou A, Kioutsioukis I, van der AR, Boersma KF, van Roozendael M, Richter A (2009) Satellite observations and model simulations of tropospheric NO2 columns over south-eastern Europe. Atmos Chem Phys 9:6119–6134

Zyrichidou I, Koukouli ME, Balis DS, Kioutsioukis I, Poupkou A, Katragkou E, Melas D, Boersma KF, van Roozendael M (2013) Evaluation of high resolution simulated and OMI retrieved tropospheric NO2 column densities over Southeastern Europe. Atmos Res 122:55–66

Zyryanov D, Foret G, Eremko M, Beekmann M, Cammas JP, D'Isidoro M, Elbern H, Flemming J, Friese E, Kioutsioukis I, Maurizi A, Melas D, Meleux F, Menut L, Moinat P, Peuch VH, Poupkou A, Razinger M, Schultz M, Stein O, Suttie M, Valdebenito A, Zerefos C, Dufour G, Bergametti G, Flaud JM (2012) 3D evaluation of tropospheric ozone simulations by an ensemble of regional Chemistry Transport Model. Atmos Chem Phys 12:3219–3240. doi:10.5194/acp-12-3219-2012