Transferring surface emissions’ heterogeneity to pollutant concentrations’ variability with a chemistry transport model

MYRTO VALARI AND LAURENT MENUT

Laboratoire de Météorologie Dynamique / IPSL, Palaiseau, France

Corresponding author address:

Myrto Valari, Laboratoire de Météorologie Dynamique,
Ecole Polytechnique 91128 Palaiseau, France.

E-mail: myrto.valari@lmd.polytechnique.fr
ABSTRACT

Grid-based chemistry-transport models are limited to horizontal resolutions of a few square kilometers. Beyond this scale explicit calculation of the turbulent flow is required in order to correctly account for chemical reactions of time-scales similar or smaller to the characteristic time-scale of turbulence. This kind of modeling is computationally too expensive to be applied over domains representing an entire urban area. However, recent human exposure studies have demonstrated the need to assess pollutant concentrations’ variability at the ‘neighborhood scale’ (i.e. of the order of a hundred meters). Here we propose a general methodology that combines existing information on the relative contribution of different emission sectors with land-use data, both provided at chemistry-transport models’ sub-grid scale, in order to disaggregate the grid-averaged emission flux into a set of source-specific components. Several concentrations are calculated under the forcing of each different emission sector resulting to concentration fluctuations around the grid-averaged value provided by the common chemistry-transport modeling procedure.
1. Introduction

The estimation of human exposure inside urban environments requires information of pollutant concentrations at local scale that is typically the scale of an administrative unit such as a census tract or a conveniently defined neighborhood (Georgopoulos et al. 2009). Regional scale chemistry-transport models (CTMs) cannot provide concentrations at such high resolution due to limitations in their parametrizations of the turbulent flow that are not adequate to represent chemical reactions of time-scales similar or smaller than the characteristic time-scale of turbulence (Vilà-Guerau de Arellano et al. 2004). Monitor data on the other hand, and especially at urban areas where surface emissions are very heterogeneous, are inadequate indicators of pollutant levels at local scales since they are directly influenced by their close environment (e.g. close to a roadway intersection, inside street canyons, residences, parks etc.) and fail to represent concentration gradients around the sources (Sapkota and Buckley 2003; Skov et al. 2001; Zhu et al. 2002). Different methods have been developed to overcome the problem. One approach is the combination of regional scale chemistry-transport modeling — to provide background pollutant levels — with local scale dispersion models to capture small scale variability of the concentration fields near emission sources (Isakov et al. 2007). However these models have limited ability to handle chemical transformations inside the reactive plume, and also obstacles such as buildings are bound to interfere to the dispersion pattern and thus need to be explicitly taken into account (Touma et al. 2006). Alternatively, spatio-temporal statistical analysis methods have been used for the interpolation of observed or predicted data to local scales (Georgopoulos et al. 2005; Christakos and Serre 2000; Bogaert et al. 2009). However those
methods risk to filter out extreme events and it is difficult to represent the sharp horizontal gradients of concentration fields near intense emission sources such as, for example, highways. Air-quality model outputs have been Monitor data have been fitted to ‘correct’ air-quality model output fields in several ways (Denby et al. October 2009): from simple kriging of model error (Blond et al. 2003) to more complex data assimilation methods (Elbern et al. 1997).

The method proposed here attempts to capture sub-grid features of pollutant concentration variability by readapting the calculation of the concentration in a grid-based CTM in order to include existing — but commonly unexploited — small scale information related to emissions and land-use data. The innovation of the approach is the suggestion that with an appropriate use of sub-grid scale data it is possible to represent the heterogeneity of surface emissions by an ensemble of emission scenarios that will drive the CTM simulation in order to create concentration fluctuations for both primary and secondary (e.g. ozone) pollutant concentrations. Thus, a CTM is used at its highest resolution (typically around 2-4km, here 3km) but at each model grid-cell emissions are disaggregated to the separate contributions of the different types of co-existing sources. Combining the intensity of each emission sector with the area of the corresponding activity we define ‘sub-areas’ of the grid-cell, where chemical transformations take place under the forcing of different emissions, such as residential, roadways, parks etc. The methodology presented here follows the same line of thought as the recent works of Galmarini et al. (2008) and Cassiani et al. (2009) since the goal is to model the sub-grid scale variability of pollutant concentrations induced by heterogeneous surface emissions without explicitly resolving the finer scale. The substantial difference is that here we attempt to simulate concentrations inside source-specific sub-grid
environments (near roads, residential, parks etc.) rather than to account for the bulk amount of variability that is generated inside the grid-cell. The advantage of this is that concentrations modeled under each emission scenario are directly connected to specific areas of the cell and therefore, more adequate for use in human exposure studies. After a short description of the applied methodology (Sect. 2) we test the ability of the method to capture sub-grid scale features of chemical reactive species on an idealized computational experiment: we compare the variability modeled with the proposed methodology with the one explicitly resolved by the same model running at a finer resolution (Sect. 3). Then we apply the computation over a real case-study between the 1st of June and the 31st of August 2006 over the regional domain centered around the city of Paris (France). Concentrations modeled separately under the forcing of traffic and residential emission sectors are compared with monitor data at sites characterized as traffic and background respectively (Sect. 4). Finally, in Section 5 we present an overall evaluation of the proposed methodology as well as a discussion on further improvements of the approach.

2. Methodology

Pollutant concentrations modeled with regional scale CTMs represent a mean value over areas of 2 to 4 square kilometers, over which emissions are considered to be homogeneous. However, over the urban environment those areas typically consist of a mosaic of emitting surfaces such as residential areas, roads, parks or other kind of sources. The chemical composition of the emitted mixture released over each one of these environments is highly
variable. Table 1 shows the average chemical composition of the emissions released by each one of the aforementioned environments calculated across the 3km×3km grid-cells of the domain covering the urban center of the city of Paris. Chemical reactions of time-scales smaller (faster) or similar to the characteristic time-scale of turbulence — responsible for the mixing of emissions inside grid-cells — modify pollutant concentrations locally (i.e. inside those micro-environments) before reactants get mixed (Krol et al. 2000; Auger and Legras 2007). These sub-grid scale features are not explicitly captured by CTMs and unless parametrized (e.g. Vinuesa and Port-Agel (2008); Vinuesa and de Arellano (2005)) they are bound to lead to errors in modeled concentrations.

Our aim is to force the CTM simulation with an ensemble of emission fluxes that represent the sub-grid scale variability of the co-existing emission sectors. This is done in two steps: the ’disaggregation’ step consists of splitting the commonly used grid averaged emission flux into relative parts representing the contribution of different emission sectors. This step requires highly resolved emission data in order to estimate the emitting activity of each emission sector inside the grid-cell. We considered four different kind of emission sectors ’residential’, ’traffic’, ’natural’ and ’other’ (the latter consists of all emission sources that are not included to the other three categories). For each considered chemical species we summed up the mass emitted per unit area and unit time from each one of the four emission sectors. The additional information in this estimation, compared to the common modeling approach, refers to the ’sector dimension’ of the emission field. Note that the described disaggregation of emissions to the contribution of different sectors is not resolved in space since all sources of the same sector included in one model grid-cell are summed up to a single value. However the disaggregation of emissions in sectors alone is not enough to
represent the desired variability in the forcing term because even if it accounts for the relative strength of each emission sector inside the cell there is still no information on the relative size of the different types of emitting surfaces. In order to correctly define the emission related to a source specific environment we need to know whether the emitted mass is released by a narrow surface (e.g. by a road) or by a wider area (e.g. a residential block). Thus, in the second step, the disaggregated emission components are scaled with coefficients representing the grid ratios covered by each different type of environment (i.e. roads for traffic emission, residential blocks for residential emissions, parks for natural emissions etc.). For example if the contribution of the traffic sector to the grid-averaged NO emission is \( E_{\text{Traffic}} \) and the area fraction of the grid surface occupied by roads is \( A_{\text{Roads}} \) then a sub-grid environment named 'Traffic' is considered, where the local NO emission forcing equals \( E_{\text{Traffic}}/A_{\text{Roads}} \). The same reasoning is applied for the estimation of the 'residential' \( E_{\text{Residential}}/A_{\text{Residential Blocks}} \), 'natural' \( E_{\text{Natural}}/A_{\text{Parks}} \) and 'other' \( E_{\text{Other}}/A_{\text{Other}} \) emission forcing at each model grid-cell.

Therefore, instead of considering that emission is homogeneous over each model grid-cell we model several concentrations, each one under the forcing of a different source term. The temporal evolution of the concentration over each different sub-grid surface reads:

\[
\frac{\partial c_i}{\partial t} + U \frac{\partial c_i}{\partial x} = - \frac{\partial}{\partial z} \left( K_z \frac{\partial c_i}{\partial z} \right) + E_i + R + Lc_i + \frac{(c_i - \bar{c})}{T_{\text{mix}}} \tag{1}
\]

where \( c_i \) is the concentration modeled under the forcing of the \( i^{th} \) emission sector. The other terms represent advection by the mean wind, turbulent advection parametrized by a gradient diffusion hypothesis, the emission term due to the \( i^{th} \) sector, source and loss terms
related to chemical transformations and deposition. The last term on the right represents an interaction between the local and the grid-averaged concentration and it is expressed by a linear relaxation of the local concentration to the local mean. The rate of the process is controlled by a time-scale $T_{mix}$. This time-scale is defined here as the ratio of a characteristic length scale and the mean local wind speed. The characteristic length is defined separately for each emitting surface depending of the spatial extend of the corresponding sector inside the grid-cell. So for narrow surfaces (e.g. roads) the mixing of the local concentration will be faster than the mixing for larger emitting surfaces (e.g. residential blocks). Under high wind speed conditions the local concentrations will tend to be mixed faster (lower $T_{mix}$) than under lower wind speed conditions (higher $T_{mix}$). A more formal definition of this characteristic time-scale should take into account the intensity of turbulence, which is responsible for the mixing at sub-grid scale (e.g. Cassiani et al. (2005)), as well as the three-dimensional structure of the corresponding source-specific environment (e.g. roads bounded by high buildings). However this kind of parametrization is not in the scope of the present study, which aims to present a more general methodology. It should be the object of a further refinement of the proposed methodology in the future though. At each model time-step the source specific concentrations are re-combined to provide the grid-averaged concentration that is transported across the domain of the simulation. We used a linear implementation of this re-combination process where source-specific concentrations are weighted with the corresponding land-use fractions: $\sum_i A_i \cdot c_i$. 

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3. Evaluation of the method based on a controlled case

a. Overall picture

The first testing of the proposed modeling is by means of CTM simulations over a controlled emissions case. CTM simulations are conducted in a modeling domain of $36\text{km} \times 36\text{km} \times 5500\text{m}$. Surface emissions are attached only to the central area of this domain. We used a random process to distribute emissions of 4 different sectors (‘residential’, ‘traffic’, ‘parks’ and ‘other’) on a 1km resolution grid over a $15\text{km} \times 15\text{km}$ area in the center of the modeling domain. We thus, impose a heterogeneous emission pattern to force the simulations (Fig. 1). The 1km spatial resolution was chosen because it corresponds to the highest resolution of CTMs. Therefore, a simulation forced by this emission input provides the finest resolution in model output possibly captured by a grid-based CTM. A first simulation is run over the 1km grid with the model running at its standard configuration. The spatial variability in the concentration fields modeled by means of this ‘reference’ simulation is explicit and it can be compared to the variability estimated with the proposed method, where simulations at lower resolutions are run under the forcing of the separate contributions of each emission sector. Model grids of 3, 6 and 12 km are used over the same modeling domain. Instead of aggregating emissions to a grid-averaged value, which would have been the case of the common modeling approach, we estimate concentrations under the forcing of the source-specific emissions following the calculation described in the previous section. At each grid-cell of the coarser resolution domains (3, 6 and 12) we estimate concentration variability as the difference between concentrations modeled over the source-specific emitting surfaces with the explicit
variability modeled by the CTM at the ‘reference’ simulation at 1km.

Meteorological conditions are also imposed for the simulations. In particular the wind speed is fixed at 1m/s across the model domain and at a constant south-west direction. Under such low wind conditions the reactants stay long enough inside each model grid cell so that sub-grid features of emission variability are transferred to modeled concentrations locally. Also this allows us to better compare model results at different resolutions because the impact of the cross-domain transport on concentrations is low compared to the influence of emissions. Boundary layer height is subjected to a diurnal cycle with a maximum of 2000m at 15:00. The boundary conditions for species concentrations are all fixed at zero except for ozone, for which a constant value of 30 ppb is uniformly assigned to the boundaries to respect typical background levels. For all resolutions and model configurations 24 hours of pollutant concentrations are simulated.

So at this stage we look to quantify what part of the variability that a CTM is able to capture at its highest possible resolution (i.e. 1km) can be represented by a model running at a coarser resolution (3, 6 and 12km) by adding some sub-grid scale features of surface emissions’ variability. Figure 2 gives a snapshot of the simulation at the 1km domain for a primary emitted species (NO\textsubscript{2}) and for a secondary pollutant (O\textsubscript{3}). Under the influence of the south-west wind emissions are directed towards the north-east part of the emission domain, where higher NO\textsubscript{2} concentrations are observed. As the flow exits the emission domain NO\textsubscript{2} concentrations decrease. Apart from that transport-related variability in modeled concentrations, local features of variability are also captured. On the other hand, as ozone is transported over the emission domain it starts being consumed for the oxydation of NO to NO\textsubscript{2}, which explains the concentration decrease towards the wind
direction. However, here also we can observe local-scale variability due to emissions’ heterogeneity. Local ozone concentration minima are found over grid-cells dominated by ‘traffic’ emissions whereas local maxima are observed over grid-cells covered by ‘parks’.

We will now focus on the simulations at the coarser grids in order to compare the variability between source-specific (sub-grid) concentrations and the one explicitly resolved at 1km. As measure of modeled variability we will be using here the difference between the concentration modeled under the residential and the traffic emission forcing for the sub-grid calculation and the difference between the concentrations modeled explicitly over grid-cells dominated by residential and traffic emissions for the 1km simulation (reference case). In order to compare model results at different resolutions we averaged spatially the source-specific concentrations modeled at the 1km grid-cells over surfaces corresponding to the lower resolution grid-cells. The results of the comparison between the different simulations are shown in Table 2. Concentrations modeled over the source-specific surfaces are given for both the explicit 1km simulation of reference and the calculation at sub-grid scale at 3, 6 and 12km resolutions. The absolute difference between concentrations estimated under the ‘residential’ and ‘traffic’ emission forcing is given for each simulation as well as the percentage of the explicitly modeled concentration variability that is captured by the proposed methodology at the lower resolutions (last column). For the comparisons with the 3 and 6km resolutions, where several grid cells are included in the 15km×15km emission domain, we present results for two different cells: those where the percentage of the explicit variability captured by the sub-grid calculation is maximum and minimum (first and second rows respectively). For the 12km resolution there is only one model grid-cell covering the entire emission domain so a unique sub-grid variability value is
Over ’residential’ surfaces, NO\textsubscript{2} and PM\textsubscript{10} concentrations are lower than those modeled over areas dominated by ’traffic’ emission. The opposite is true for ozone concentrations. These remarks are general and apply to both explicit calculations and sub-grid estimates and to all resolutions. For the primary emitted species (NO\textsubscript{2} and PM\textsubscript{10}) this is directly connected to the fact that higher emissions are released over roadways than over residential areas. On the other hand ozone is consumed over roadways, where NO emission is high, for the oxidation of NO to NO\textsubscript{2}, which explains the low ozone concentrations modeled under the forcing of the ’traffic’ emission sector. An other issue is that the difference in concentrations modeled under the forcing of the ’residential’ and the ’traffic’ emission sectors at 3, 6 and 12km with the proposed method is very similar for all the selected grid-cells and depends very little on the resolution. This is not the case for the explicitly modeled concentration variability over the corresponding emitting surfaces. This reflects the fact that over the 1km grid the transport of emissions inside the block of cells corresponding to the coarser resolutions’ surfaces is explicit. Therefore the relative position of the emission surfaces over these areas matters for the calculation. This is not the case for the sub-grid estimate since the mixing of emissions inside the grid-cell is parametrized as a function of the wind-speed and the area fraction occupied by each emission sector. In the controlled simulation analyzed here the wind speed is constant and the occupation fractions vary a little between grid-cells. It is therefore not surprising that only a small difference is observed in modeled variability between grid-cells.
b. Time-series at selected grid-cells

In this section we focus on selected model grid-cells in order to study more in detail the simulation results under the forcing of each one of the four emission sectors. Since it was shown that modeled variability little depends on both the grid-cell and grid resolution we present the results from the simulation over the 12km resolution grid (Fig. 3). Some common features are observed for the three chemical species under consideration: first of all it is shown that the mean concentration calculated from the 1km$^2$ grid-cells over surfaces of 12km$\times$12km are very close to the grid-averaged concentration modeled directly at the 12km resolution grid from the concentrations estimated under the source-specific emission forcing. This remark shows that the recombination of the source-specific concentrations to a grid-averaged value does not alter the commonly applied calculation. On the other hand the source-specific concentrations represent correctly the deviation from the mean value if they are compared to the explicitly modeled variability. A certain underestimation of the overall variability is observed at all cases which is due to the fact that only variability in the emission forcing is taken into account with the sub-grid implementation whereas the explicitly resolved variability at 1km also considers the relative position of the emitting surfaces inside the 12km$^2$ area. The concentration related to the ‘residential’ emission is in all cases attached to the mean value. This is due to the fact that the larger part of the emission domain is covered by surfaces releasing ‘residential’ emissions and therefore, there is little difference between the grid-averaged and the ‘residential’ emissions forcing leading to similar concentrations.

However, if the concentrations forced by each emission sector are compared to the explicit
calculation at 1km it is shown that the deviation from the grid-averaged value is represented accurately. Also, this suggests that the separation of the forcing into emission sectors is correctly implemented into the CTM. For ozone the highest concentrations are modeled over the 'other' emission surfaces. This sector has been added as a separate forcing in the controlled emissions model run but it does not represent a specific source of emissions but rather the contribution of several point sources such as industrial stacks etc. Because these sources are scarce inside the highly urbanized center of the cities the mass emitted from the 'other' sector is significantly lower than the emissions from the dominant sectors (traffic and residential). Also, emissions released by the 'natural' sector are low compared to those emitted by the dominant sectors. A rounded estimate of the relative rate of NO\textsubscript{x} emission in each sector is 5 : 25 : 75 : 100 for the 'other', 'natural', residential' and 'traffic' sectors respectively. These ratios are directly reflected in modeled concentrations: for NO\textsubscript{2} modeled concentrations are higher for sectors of high NO\textsubscript{x} emissions and lower over surfaces of low NO\textsubscript{x} emission. On the other hand, ozone concentrations are higher there, where NO\textsubscript{x} emission is low and lower close to high NO\textsubscript{x} emission. These remarks apply to both the 1km explicit simulation and the sub-grid calculation over the 12km resolution grid.

c. Sensitivity to the implementation of the sub-grid mixing

Concentrations modeled under the forcing of each separate emission sector are transported across the grid exactly as the mean concentration, by means of grid-averaged fields calculated locally for each grid-cell (i.e. mean wind, vertical diffusion coefficient, dry
deposition velocity). Grid-to-grid concentration gradients are calculated as the difference between source-specific (for the local) and grid-averaged (for the adjacent) grid-cells. Thus, through the transport process the source-specific concentrations interact with the 'mean model'. However, this interaction occurs at the grid scale. The interaction between source-specific concentrations and the mean model at sub-grid scale is implemented by the last term on the right of Eq. 1. Here, we consider that the source-specific concentrations get diluted into the local average concentration over an 'interaction area' whose extent depends on the size of the corresponding emitting surface. The time-scale of the process depends also on the wind speed, which we consider here, responsible for the mixing. This implementation is usually referred to as relaxation of the local concentration towards the local average and its time-scale is expressed as $T_{mix} = CT$, with $T$ the ratio of the turbulent kinetic energy and the mean turbulent energy dissipation rate and $C$ a constant of proportionality (see Cassiani et al. (2009)). The CTM used for the present study does not provide any measure of the turbulent time-scale in the horizontal dimension, we therefore used the mean wind speed in order to deduce the time-scale of the relaxation process. Also, our implementation implies that the proportionality constant is one.

Figure 4 shows the sensitivity of modeled variability to the constant of proportionality — the wind speed is constant throughout the simulation and does not influence the calculation of the time-scale of the sub-grid mixing. The difference in NO$_2$ concentration between the 'traffic' emission sector and the 'mean' model changes by 12% when $C$ varies with in a range of 0.1 to 2. As shown in Fig. 4 for $C$ around 1 model variability is independent on $C$. However this result is obtained under controlled meteorological conditions with constant wind. We do not imply, by no means, that the same stability in
model variability would be observed under stronger turbulent conditions.

d. Sensitivity to other meteorological parameters

As the air flow is transported over the emission domain the concentration of primary emitted species will tend to increase because of accumulation. On the other hand, ozone concentration will decrease in the direction of transport due to depletion close to NO emissions. Ozone concentrations will start to increase again after the flow exits the emission domain to reach background levels (we consider here the case of limited photochemical activity). These spatial gradients in the concentration fields risk to overshadow the variability in pollutant concentrations induced by local-scale emission heterogeneity. In order to capture these small-scale features of variability we used constant wind of very low speed (1m/s). Under such conditions concentration varies slowly with transport and variability induced by local emissions dominates. However, we studied the response of the implemented calculation to different meteorological conditions, and in particular its sensitivity towards the wind speed and the boundary layer height (BLH). We considered the same emission domain over a 3km×3km resolution grid from which a non-ractive tracer was released at different rates from each one of the four emission sectors. Figure 5 shows the results of this study: modeled variability decreases as the wind speed becomes higher (Fig. 5 (top)); emissions are rapidly mixed and the induced variability is homogenized. On the contrary the variability of the tracer’s concentration at the surface level increases as the BLH becomes higher (Fig. 5 (bottom)). In the CTM used here, the vertical diffusion coefficient (K<sub>z</sub>) at the first vertical
level is deduced from the BLH so that high boundary layers are associated with strong upward drafts (Vautard et al. 2001). Stronger upward diffusion over heterogeneous emissions results in higher tracer variability at surface level, which can be explained by the fact that vertical motion overweighs horizontal transport under such conditions. Vertical diffusion preserves variability while horizontal transport tends to suppress it. The important point here, is that both estimates of variability — the explicit modeling at 1km and the proposed methodology at 3km resolution — respond in the same manner to changes in the meteorological parameters considered in the study.

4. Application of the method into realistic atmospheric conditions

Based on model-to-model comparisons we showed that by splitting the grid-averaged emission forcing into separate terms representing the sub-grid scale contribution of dominant emission sectors we are able to capture the largest part of the variability in pollutant concentration fields that had been explicitly resolved by the model running directly at the finer resolution. This kind of modeling is very convenient since it provides information on small-scale features of pollutant concentration variability without increasing model resolution — and consequently the time of the computation.

At this stage we will apply the calculation on a real case study. The CHIMERE model (Rouil et al. 2009) is run over a 3km×3km resolution grid over a domain of 160km×130km (53×43 grid-cells) covering the greater Paris area. Hourly averaged pollutant
concentrations are modeled for the 3-months period from June 1st to August 31st 2006. The separation of emissions into source-specific contributions used to force the CTM simulation and create sub-grid scale concentration variability is based on data provided by the local emission inventory of AIRPARIF at 1km resolution (Vautard et al. 2003). From this inventory we retrieved hourly averaged emission rates per species and per emission sector. Emissions are categorized into 11 principal sectors covering activities such as industrial and combustion, transport, agriculture and nature emissions, further divided into sub-categories leading to a total number of 44 types of emission sources. Here we aggregated these sectors into the four classes presented in the previous section (i.e 'traffic', 'residential', 'natural' and 'other'). Figure 6 shows the 3km grid-averaged emission rates of NO\textsubscript{x} as well as the relative contribution of the 'traffic' and 'residential' sectors over the greater Paris region (left panels). Then emission rates per sector were treated to represent grid ratios using land-use based area fractions as relative weight coefficients. These coefficients were deduced from land-cover data initially organized in 42 categories at a 100m resolution (CORINE Land Cover) and then aggregated into four land-use classes — 'roadways', 'residential areas', 'parks', 'other' — each one corresponding to one of the 4 emission sectors. The emission input used to force the 'residential' and 'traffic', source-specific simulations are shown on the right panels of Fig. 6. The calculation over source-specific emission surfaces is applied only on the central area of the domain, where emissions released from the city of Paris are the most heterogenous.
Pollutant concentrations modeled under the forcing of the different emission sectors are linearly recombined to a mean concentration using as weight coefficients the source-specific grid-ratios associated to the corresponding emission sector. This mean concentration was compared to the grid-averaged concentration modeled with the CTM at its 'standard' configuration (i.e. forced by the grid-averaged emissions). The histograms of Fig. 7 are calculated over all the grid-cells, where the sub-grid calculation was applied, and for all the hours of the 3-months' simulation. The distributions of the differences between the two calculations are given for three chemical species (i.e. O$_3$, NO$_2$ and PM$_{10}$). Ozone calculated as the mean of the concentrations modeled under the separate forcing of each emission sector is always higher than ozone modeled under the 'standard' grid-averaged emission scenario. Generally CTM’s underestimate ozone over cities because NO emissions are averaged over heterogeneous surfaces such as roadways — with very high NO emission — and other surfaces where NO emission is significantly lower (e.g. residential emissions) (Seinfeld and Pandis 1998). If the grid-averaged NOx emission is high enough to bring NOx/VOC ratio over the threshold value between the two chemical regimes too much ozone will be consumed at the fast NO+O$_3$ → NO$_2$+O$_2$ reaction over the entire grid-cell area. The strictly positive difference between sub-grid and grid-averaged calculations imply that this non-linear aspect of ozone chemistry can be better represented by modeling separately ozone concentrations under the emissions of different environments rather than average emissions beforehand. The distribution of NO$_2$ differences is symmetrical to the one of ozone differences with negative values. This reflects the same effect 'seen' from the
point of view of the product of the NO oxidation. Less NO is converted to NO\(_2\) when concentrations are modeled first over each separate emission surface and then averaged than directly under the grid-averaged emission scenario. The distribution of PM\(_{10}\) differences implies that the sub-grid calculation little affects the mean value of its concentration. Most of the time differences are very close to zero, with a small spread on both positive and negative values. PM\(_{10}\), as a family of chemicals, is much less reactive than gases such as NO\(_x\) or ozone. The mean concentrations modeled either way are therefore very close one to the other.

b. Comparison with surface measurements

At this section we will focus on specific model grid-cells in order to compare the variability in pollutant concentrations captured by the sub-grid calculation with the one measured by monitors located at environments characterized by different types of pollution. More precisely we will compare concentrations modeled under the ’residential’ and ’traffic’ emission forcing with monitor data collected at ’background’ and ’traffic’ sites respectively. The specific model grid-cells that will be used for this comparisons are shown in Fig. 8. Also Table 3 gives the area fractions of the grid-cells occupied by the different types of surfaces. Three different monitors are located inside the central model grid-cell (i.e. the background monitors BG1 and BG3 and the traffic monitor TR1). The surface of the corresponding grid-cell is occupied by roadways, residential area, parks, and other kind of surfaces by approximately 5, 35, 53 and 7 % respectively.

When both ’background’ and ’traffic’ monitors of a specific pollutant are located inside the
same model grid-cell we compare directly modeled and measured variability. Unfortunately this is possible only for NO\textsubscript{2} with BG1 and TR1 monitors. Ozone is not measured at any traffic station and PM\textsubscript{10} are only measured at areas where no PM\textsubscript{10} background monitor exists nearby. In the later case we show results at different model grid-cells. Time-series of modeled concentrations and monitor data are given in Fig. 9. On the left panels we compare concentrations of the three pollutants modeled under the forcing of the residential emission sector with data measured at 'background' sites inside the corresponding model grid-cell. For all three species the concentration modeled under the 'residential' emission forcing is very close to the grid-averaged concentration. Residential areas are much larger than the total grid surface covered by roadways. So even if the mass per unit area and unit time is lower for 'residential' emissions than for 'traffic' emissions it is still the dominant component of the grid-averaged emission released over the grid-cell surface. Nevertheless, the deviation from the grid-averaged value observed for the 'residential' concentration brings model results closer to the background measurement than the commonly modeled grid-averaged concentration for all three chemical species. For primary emitted pollutants such as NO\textsubscript{2} and PM\textsubscript{10} the concentrations modeled under the residential emission scenario are lower than the grid-averaged value. On the other hand, higher ozone concentrations are modeled over residential areas than with the grid-averaged emission forcing. Under the 'traffic' emission forcing modeled concentrations deviate a lot from the grid-averaged value, with differences that may exceed 30 \(\mu\text{g/m}^3\) for all three pollutants. For PM\textsubscript{10} and NO\textsubscript{2} comparisons with monitor data show that modeled concentrations are realistic. NO\textsubscript{2} and PM\textsubscript{10} concentrations are higher under the 'traffic' emission scenario but less ozone is modeled compared to the grid-averaged simulation. These observations show
that by separating the emission forcing per sectors it is possible to represent the variability in the chemical regimes determined by the VOC/NO\textsubscript{x} ratios (Sillman and H.Dongyang 2002). Close to roadways (VOC/NO\textsubscript{x} emission ratios <1), ozone is consumed for the NO oxidation to NO\textsubscript{2} leading to high NO\textsubscript{2} and low ozone concentrations. Over residential areas, where VOC/NO\textsubscript{x} emission ratio is high, organic peroxides are used for the oxidation of NO to NO\textsubscript{2} instead of ozone. With the commonly used grid-averaged approach this difference in not captured and ozone titration is often overestimated by models over highly urbanized areas. Here the separation in the emission forcing leads to realistic concentrations over the different emission surfaces without perturbing the grid-averaged calculation.

5. Discussion

In this paper we propose a novel methodology for the computation of pollutants’ variability between urban environments characterized by very different types of surface emission. Very often, over cities these environments happen to be too close one to the other that even the finest resolution of chemistry-transport models is incapable to resolve the variability imposed to pollutant concentrations. Increasing the model resolution in order to represent explicitly the scale of emissions’ variability is computationally too expensive and also increases model uncertainties, especially considering the difficulty to define accurately the location and rate of emissions from various sources, many of which being mobile (Russell and Dennis 2000). For this reason we propose an alternative computation that uses only certain finely resolved data in order to reconstruct a limited number of ’sub-grid environments’ representing the most commonly encountered types of urban pollution. We
examined the possibility to represent the variability of concentrations at sub-grid scale by modeling separately concentrations inside each sub-grid environment. We evaluated the method first by trying to reproduce the concentration variability modeled explicitly at 1km (maximum CTM resolution) with simulations run over a coarser grid. Apart from a certain underestimation of the overall variability, which is not surprising since our calculation is not explicit, the method was shown capable to represent correctly the difference in concentrations over heterogeneous emission surfaces. The method was also tested on a real case-study and we compared the concentrations modeled inside the considered sub-grid environments with measurements representing different types of pollution. The information added to the CTM’s grid-averaged concentration by means of the implementation of the sub-grid calculation was in good agreement with the variability captured by the monitors. We have to note here that the comparison with measurements presented in this study is not exhaustive (e.g. no traffic monitor sites for ozone, no traffic and background monitors inside the same grid-cell for PM$_{10}$). This comparison should be seen more as an example aiming to compare the order of magnitudes of modeled and observed variability rather than as an exhaustive model validation. More generally, the comparison between model and monitor data in the context of concentration variability is not straightforward (Ching et al. 2006; Gelfand et al. 2001). It is beyond the scope of the present study to propose a framework that would make possible such a comparison but it is worth noticing that the modeling of pollutant concentrations inside sub-grid environments is a possible way to overcome a long standing problem of comparing model concentrations with measurements of different spatial representativity. The success of the proposed implementation depends strongly on the way the sub-grid environments are defined. This includes the selection of
the kind of highly resolution data that will be used and also on their quality. For example a point that needs to be refined in the implementation presented here is the parametrization of the sub-grid mixing. This parametrization should take into account additional information concerning the three-dimensional structure of the corresponding environment (e.g., buildings’ height, roadways’ width etc.). It is possible to derive such a parametrization from campaign measurements focusing on near-roads concentration gradients or local models that simulate the dispersion of emissions from point sources.
REFERENCES


Galmarini, S., J.-F. Vinuesa, and A. Martilli, 2008: Modelling the impact of sub-grid scale emission variability on upper-air concentration. *Atmospheric Chemistry and Physics, 8*, 141–158.


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<table>
<thead>
<tr>
<th></th>
<th>NH$_3$</th>
<th>SO$_2$</th>
<th>NO$_x$</th>
<th>VOCs</th>
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<td>12%</td>
<td>27%</td>
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<tr>
<td>Traffic</td>
<td>0</td>
<td>0</td>
<td>76%</td>
<td>23%</td>
</tr>
<tr>
<td>Natural</td>
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<td>0</td>
<td>20%</td>
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<tr>
<td>Other</td>
<td>0.4 %</td>
<td>30%</td>
<td>31%</td>
<td>38.6%</td>
</tr>
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Table 1. Relative mass emitted per chemical species with respect to the total emitted mass by the corresponding emission sector (%). The sector denoted as ‘Other’ refers to emissions from sectors that are not included into the other three categories.
<table>
<thead>
<tr>
<th>NO₂</th>
<th>CTM 1km explicit</th>
<th>CTM 3, 6, 12km sub-grid</th>
<th>Resid</th>
<th>Traffic</th>
<th>Δₐₓ</th>
<th>Resid</th>
<th>Traffic</th>
<th>Δₐᵟ</th>
<th>%</th>
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<tbody>
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<td>3×3 km²</td>
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<td>4.55</td>
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<td>12×12 km²</td>
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<td>-0.63</td>
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<td>PM₁₀</td>
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Table 2. Comparison between the explicit variability of NO₂, PM₁₀ and O₃ concentration modeled at 1km resolution over ‘residential’ and ‘traffic’ emission surfaces and the variability estimated under the ‘residential’ and ‘traffic’ emission forcing at 3, 6 and 12km grid resolution. For both the explicit and sub-grid calculations the first two columns show modeled concentrations over the corresponding surfaces (or emission forcing), while the third column is represents the absolute difference between the later concentrations, as a measure of the variability. The last column of the table is the percentage of the explicitly modeled variability with the 1km reference simulation that is capture by the sub-grid estimation.
<table>
<thead>
<tr>
<th>Model cell Station</th>
<th>Roadways (%)</th>
<th>Residential (%)</th>
<th>Parks (%)</th>
<th>Other (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>BG1, TR1</td>
<td>4.8</td>
<td>35.2</td>
<td>53.3</td>
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<td>BG2</td>
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<td>6.6</td>
<td>0.0</td>
</tr>
<tr>
<td>TR2</td>
<td>3.6</td>
<td>81.3</td>
<td>0.0</td>
<td>15</td>
</tr>
</tbody>
</table>

**Table 3.** Fractions of occupation of model grid-cells by different types of surfaces (land-use). The location of these cells on the model grid is shown in the Fig. 8.
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1. Controlled case emitting surfaces spread over the 1km resolution grid. Four different types of emitting surfaces are considered 'Residential', 'Traffic', 'Natural' and 'Other'.

2. Surface concentrations of (left) NO$_2$ (ppb) and (right) O$_3$ (ppb) at 14:00 UTC estimated by the controlled case simulation over the 1×1 km$^2$ grid. Note that the white color in both maps corresponds to the boundary condition levels (0ppb for NO$_2$ and 30ppb for O$_3$).

3. Time series of (top) O$_3$, (middle) NO$_2$ and (bottom) PM$_{10}$ modeled (left) explicitly at 1km resolution and (right) calculated with the proposed methodology under the separate forcing of the four emission sectors at 12km.

4. Difference in NO$_2$ concentration modeled under the forcing of the 'traffic' emission sector and the local mean as a function of the proportionality constant ($C$) formally used to define the time-scale of the sub-grid mixing process.

5. Difference between the maximum and minimum concentration of a tracer gas estimated inside surfaces of 3km×3km at 12:00 UTC as a function of (top) the surface wind speed and (bottom) the boundary layer height. Concentration variability is calculated in two ways: explicitly at 1km and directly at 3km×3km by separating the emission forcing into the contribution of four dominant emission sectors.
Emission input used for the CTM simulations at 3km×3km resolution grid over the city of Paris. Left panels: (top) the grid averaged NOx emissions used for a 'standard' CHIMERE model simulation, (middle) the 'traffic' sector contribution to the total mass emitted per km² and per hour, (bottom) the 'residential' sector contribution. Right panels: emission forcing for the source-specific simulations. (Top) Grid-averaged emission, (middle) 'traffic' emission forcing, (bottom) 'residential' emission forcing.

Difference (%) between the mean surface concentrations of ozone, NO₂ and PM₁₀ averaged over the 4 source-specific concentrations and under the 'standard' grid-averaged emission forcing.

3km resolution grid over the city of Paris, showing the locations of 'background' (BG) and 'traffic' (TR) monitors and the corresponding model grid-cells.

Time-series of surface pollutant concentrations modeled under the separate forcing of different emission sectors and with the 'standard' grid-averaged emission input. Also the time-series of monitor data at 'background' and 'traffic' stations are compared to model results. Comparisons at 'background' sites are given on the left panels and comparisons at 'traffic' stations are given on the right panels.
Fig. 1. Controlled case emitting surfaces spread over the 1km resolution grid. Four different types of emitting surfaces are considered ‘Residential’, ‘Traffic’, ‘Natural’ and ‘Other’.
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Fig. 6. Emission input used for the CTM simulations at 3km×3km resolution grid over the city of Paris. Left panels: (top) the grid-averaged NO\textsubscript{x} emissions used for a ‘standard’ CHIMERE model simulation, (middle) the ‘traffic’ sector contribution to the total mass emitted per km\textsuperscript{2} and per hour, (bottom) the ‘residential’ sector contribution. Right panels: emission forcing for the source-specific simulations. (Top) Grid-averaged emission, (middle) ‘traffic’ emission forcing, (bottom) ‘residential’ emission forcing.
Fig. 7. Difference (%) between the mean surface concentrations of ozone, NO$_2$ and PM$_{10}$ averaged over the 4 source-specific concentrations and under the 'standard' grid-averaged emission forcing.
Fig. 8. 3km resolution grid over the city of Paris, showing the locations of 'background' (BG) and 'traffic' (TR) monitors and the corresponding model grid-cells.
Fig. 9. Time-series of surface pollutant concentrations modeled under the separate forcing of different emission sectors and with the ‘standard’ grid-averaged emission input. Also the time-series of monitor data at ‘background’ and ‘traffic’ stations are compared to model results. Comparisons at ‘background’ sites are given on the left panels and comparisons at ‘traffic’ stations are given on the right panels.