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## Addressing the issue of exposure to primary pollution in urban areas: application to Greater Paris

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#### ARTICLE INFO

### ABSTRACT

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This work aims to highlight the importance of refining the calculation of exposure to primary pollutants in dense urban areas, by taking into account both pollutant concentration heterogeneity at the street scale, and individual mobility during the day. To address this issue, we have set up an urban modeling chain based on the OLYMPUS and CHIMERE models, and offering innovative statistical work on simulation data. OLYMPUS is an emission modeling system that produces mobility matrices for individuals in a city through an activity-based approach of the travel demand. CHIMERE is a chemistry-transport model that uses anthropogenic and biogenic emissions, meteorological fields and large-scale chemical fields to produce hourly pollutant concentration fields in a given area. A statistical post-processing of CHIMERE output data has been developed to create sub-grid variability for  $NO_2$  and  $PM_{10}$ concentrations - mainly related to the proximity of busy roads, and its results have been crossed with daily mobility matrices for every citizen, in order to refine our estimate of the individual exposure budget. This urban modeling platform has been implemented over Greater Paris. The results show that such approach considerably expands the range of individual exposure values and raises the question of recurrent exposure of city dwellers to very high levels of pollution, even during reduced periods of time during the day. They also introduce the issue of environmental inequalities linked to practices and constraints of dwelling place, mobility and work, which need to be apprehended by new modeling approaches.

Abbreviations and acronyms : CHIMERE, A multi-scale chemistry-transport model for atmospheric composition analysis and forecast; COPERT, Computer Programme to calculate Emissions from Road Transport; CTM, Chemistry-transport model; NOx, Nitrogen Oxides; NO2, Nitrogen Dixide; NMVOC, Non Methanic Volatile Organic Compound; O<sub>3</sub>, Ozone; OLYMPUS, integrated air pollutant and GHG urban emissions model; PM10, Particulate matter (< 10 µm); PM2.5, Fine particles (< 2.5 µm); WHO, World Health Organization.

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### 1. Introduction

From the 1990s, air quality monitoring in Europe was very strongly structured around field observations, through the operational implementation of measurement networks centered on urbanized areas. Since measurements are based on normalized methods, their uncertainty is highly constrained (15% maximum for NO<sub>2</sub>, 25% for PM<sub>10</sub>, (2010-2050 Decree of the 21<sup>st</sup> October 2010, the 2008/50/CE and the 2004/107/CE directives) and the literature considers that air quality monitoring networks today can estimate the atmospheric content of these pollutants with high precision. Based on these measurements as validation parameters, the Eulerian Chemistry-Transport Models (CTMs) developed during the 1990s and 2000s made it possible to spatially supplement our knowledge of air quality. Since then, many modeling studies conducted as part of environmental research were intended to support decision making and to provide a quantification of the effects of the technological control of pollutant emissions on air quality, in the context of both the European regulatory directives and the regional plans for the protection of the atmosphere (Coll et al., 2009; Thunis et al., 2007). However, a collective consciousness of urban populations is emerging today around the health risks of atmospheric pollution. It follows repeated reports from institutions such as the WHO on the dangerousness of combustion particles (Pascal et al., 2014; WHO, 2016) and the recent classification of urban air pollution as a proven carcinogen (Loomis et al., 2013; Pope III et al., 2002; Raaschou Nielsen et al., 2016), but it also comes from the increase in the frequency of chronic respiratory diseases in the population of urban areas (Cesaroni et al., 2013; Perez et al., 2013). In addition, the inhomogeneity of urban exposure - strongly increased in direct proximity to the traffic lane, especially when the roads are congested or when the buildings favor the confinement of the air - is now recognized in the literature (Hoek et al., 2002). Epidemiological studies have been able to highlight the phenomenon of "contrasts of exposure", which is manifested by the deterioration of the state of health of populations living near road traffic and by the production of environmental inequalities (Deguen et al., 2015). Such findings strongly question the townspeople about their own exposure. In this context, research studies should set up a more thorough analysis of air quality issues, giving access to exposure with a level of refinement that tends towards the individual. In particular, the health benefits that can be expected from the implementation of policies must be the main drivers of their evaluation. However, our traditional approach to exposure raises methodological issues. Indeed, epidemiological studies so far have been based on spatial interpolation of measurement data at the population's residence points using Land Use Regression models, or by cross-referencing data from modeled maps of air quality and population density (Cesaroni et al., 2013), but none of them could take into account the mobility of individuals, which leads to an erroneous assessment of the exposure of the population and hence, of the induced health effects (Dewulf et al., 2016; Gurram et al., 2019; Schmitz et al., 2019). In addition, the CTMs - which are likely to feed these studies operate on a scale that does not take into account the effect of immediate proximity to sources, nor the local processes of pollutants accumulation in dense built-up areas. For these reasons, CTM outputs have always shown better comparability to measurements made in the urban background, away from the direct impact of emission sources (Valari and Menut, 2008), and do not compare well with data obtained in the core of the city, along main roads and in confined source areas. Yet it is in these dense urban spaces and close to the sources of pollution that townspeople live and move around along the day. Not considering such reality may distort our estimate of individuals exposure to pollutants. In particular, (Thompson et al., 2014) have shown that large-scale modeling tends to overestimate the benefits of reducing pollutants in cities.

Recently, many research efforts have been engaged to remove the technical locks of urban air quality modelling at the street scale. They include the coupling of CTMS with Gaussian dispersion or Computational Fluid Dynamics models to represent pollution flows in the urban area (Hülsmann et al., 2014; Kim et al., 2018; Pepe et al., 2016; Stein et al., 2007; Yuan et al., 2014). Although it is acknowledged that NOx and PM concentrations levels at the street scale are described in a more consistent way, the set-up of such models needs a large amount of refined specific data such as building geometry, local dynamics or precise mobile emissions that can be limiting, especially when modeling air quality in canyon streets (Hülsmann et al., 2014). Furthermore, computational time can be important over large domains and long time periods, as a bigger number of iterations would be needed to reach steady state (Kim et al., 2018). As alternatives, land-use regression models combining measurement data and predictors variables (accounting for landuse, traffic intensity, geographical typology and population data) have been recognized to provide a fair representation of pollutant dispersion in urban areas, while requiring less detailed input data than dispersion models (Hock et al., 2008). However, the lack of information on accurate local traffic information tends to limit their relevance for calculating fine-scale exposure (Beelen et al., 2013). Some authors have developed alternative approaches proposing to recreate the fine-scale variability observed in the measurements within the CTM meshes. These so-called downscaling approaches are based on statistical methods using proxies in order to recreate - after a learning phase - realistic concentration gradients on a finer scale than that of the CTM simulation (Chourdakis et al., 2016; Kuik et al., 2017). Their level of complexity and their prognostic or diagnostic character may vary, and their results are by structure less precise than CFD models, but they constitute a quick and simple alternative to the set-up of finescale urban models. In parallel, a growing body of several air quality studies have been dedicated to estimate punctual people exposure to air pollutants in the different indoor and outdoor urban micro-environments, as well as average chronic exposure of townspeople to pollution during their lifetime (Arvanitis and Moussiopoulos, 2006; Int Panis et al., 2010; Yang et al., 2015).

Our works are in line with these efforts around a better estimation of individual exposure to urban air pollution. More precisely, this paper aims to explore the impact of new approaches to urban modeling, on the representation of daily exposure to air pollutants in dense urban spaces. Our works are based on a new modelling platform, designed around the OLYMPUS emission model (Elessa Etuman and Coll, 2018), the CHIMERE air quality model (Menut et al., 2013), statistical downscaling of the CTM outputs and dynamic exposure calculations. The emission and air quality modelling step is described in section 2. After running air quality modelling with the CHIMERE model, we perform a series of statistical treatments of the model outputs to produce next-generation exposure diagnoses that include both urban heterogeneity and mobility. These works include statistical refinement of CHIMERE output concentration fields at the sub grid scale, as well as the consideration of the time-space exposure budget of city-dwellers. The added value of such an approach is presented and discussed here through the comparison of standard / improved air quality and exposure simulation in the Greater Paris. Our purpose is to show that not taking into account urban specificities (such as local concentration gradients) and individual mobility may underestimate the risk linked with exposure to pollutants in dense metropolises.

### 2. Configuration and set up of the OLYMPUS and CHIMERE models

We have implemented our integrated urban air quality modeling platform at the regional scale over France and for the whole year 2009, which is quite representative of the last decade in terms of air quality on the French territory. In particular, it includes an

intense episode of winter pollution that is linked to the installation of a high-pressure situation across Europe at the beginning of January. A second particulate pollution event is observed in the spring (April 3-6), as well as three photochemical episodes (NO<sub>2</sub>, O<sub>3</sub>) later in the year (https://www.airparif.asso.fr/alertes/historique). Our focus area is the Ile-de-France region, which is densely urbanized since it contains the metropolis of Greater Paris, home to approximately 12 millions inhabitants.

As shown in Fig. 1, the exposure modeling process is performed in 4 steps. The first step is the generation of an emissions inventory based on the activities of individuals living in a territory, managed by the OLYMPUS tool. The second step is to diagnose air quality using the CHIMERE 3D model, which provides hourly concentration fields of pollutants at the kilometric scale. In the third phase, a statistical downscaling module allows refining the air quality diagnosis by creating sub grid concentration gradients within the urban space. Finally, the intersection of citizen urban trips and fine-scale air quality maps makes it possible to consider the dynamic exposure of all individuals in the population.



Fig. 1. Organization of the modelling chain

#### 2.1 OLYMPUS set-up

Our simulation work started with the implementation of the OLYMPUS tool (http://www.lisa.u-pec.fr/instruments/?id=1295). OLYMPUS is an emission model that relies on the statistical representation of individuals' energy consumption practices in a territory. It allows to assess - at an individual level - the impacts of the urban form and of the organization of public transport on mobility, on energy consumption, and ultimately on the emissions of greenhouse gases and air pollutants. OLYMPUS requires as main input the parameters influencing urban organization (mostly urban morphology, population density, employment area and networks) but also survey data relating to mobility practices, equipment (vehicles, heating appliances) and to the regional energy mix. All of this data is used to produce a diagnosis of the transport and energy demand in the studied area, as well as a spatialized inventory of primary air pollutants: nitrogen oxides, fine particles, carbon oxides, volatile organic compounds and sulfur dioxide. OLYMPUS was then successfully implemented in the Île-de-France region, a territory which has an increasing density of activities towards a large urban city core. A fully detailed presentation of the operating principles of the model and its implementation on Îlede-France can be found in Elessa Etuman and Coll (2018). For the record, the OLYMPUS configuration and simulation steps can be summarized as follows. The 2009 demographic situation of the Paris region was modelled via a statistical approach based on conditional probabilities, using land use information and socio-demographic surveys from the national institute of statistics and economic studies (https://www.insee.fr/fr/accueil). Subsequently, the mobility of each individual of the synthetic population was simulated using 1) an activity-based model for the generation of the mobility demand, 2) a gravity approach for the spatial allocation of activities and 3) the allocation of transportation demand on the road network, using a short-path approach taking into account network congestion. For these steps, we used as input the regional mobility practices provided by the Global Transport Survey of Île-de-France (STIF 2012). In parallel, the energy demand of the territory was estimated from representative unit energy consumption rates for households and employees, according to statistical household practices and building properties. Finally, the emissions associated with all modeled activities were calculated using the methods recommended by the European Environment Agency (European Environment Agency, 2013). In the end, OLYMPUS provides a population mobility matrix, timed vehicle fluxes on all road axes and an emission inventory from the energy-consuming activities described above, spatialized on the 1x1km<sup>2</sup> computation grid. The emissions produced by OLYMPUS in the Île-de-France region for the year 2009 have been deeply analyzed and validated by comparison with other institutional emission inventories available on this area (Elessa Etuman and Coll, 2018). With regard to the model transferability, a study was carried out in parallel on the French Pays-de-la-Loire region, which demonstrated the ability of OLYMPUS to simulate mobility fluxes in a rural territory where multiple urban centers have developed. The main results obtained on these two regions are described at length, illustrated and compared in the companion article thereof (Elessa Etuman et al., Same session), and they demonstrate the strong potential of OLYMPUS for the restitution of the geography of traffic flows.

#### 2.2 CHIMERE set-up

CHIMERE is a chemistry-transport model dedicated to the regulatory forecast of air quality but also to research based on the exploitation of past or prospective situations, and which application scales range from urban air quality with a resolution of 1-2 km to hemispheric issues with a resolution of 100-200 km (available at http://www.lmd.polytechnique.fr/chimere/). As part of the French regulatory missions for monitoring and forecasting air quality at the regional, national and European scales, the model is implemented daily and continuously evaluated by comparison with network measurement data for nitrogen oxides, particulate matter, CO and SO<sub>2</sub>.

In the model, the calculation of air pollutant concentration levels at each grid point is based on the basic principle of mass conservation, which results from the budget of emissions, physical and chemical processing in the atmosphere as well as advection, diffusion and deposition processes. CHIMERE uses the reduced chemical mechanism MELCHIOR2 that was designed for the regional production of oxidants on a regional to continental area and takes into account 44 species (including 19 organic compounds) and 120 gaseous reactions. The aerosol module represents primary particles, biogenic and anthropogenic secondary organic aerosol species, sea salts, dust and water, all distributed into 9 size bins ranging from 40 nm to  $10\mu m$ .

In this work, CHIMERE was first implemented at the continental (resolution  $0.68^{\circ} \times 0.46^{\circ}$ ) and national (resolution  $0.205^{\circ} \times 0.135^{\circ}$ , about 15 x 15km<sup>2</sup>) scales to provide chemical boundary conditions for the Île-de-France target study area (see domains in Fig.2). Such a nesting configuration makes it possible to better represent the major European urbanized areas, as well as the emissions from the North Sea maritime corridor, which have an impact on the composition of the background atmosphere of the French territory during the period of study - and especially during the anticyclonic situation of January 2009. Subsequently, CHIMERE was run at the highest reasonable resolution ( $1x1km^2$ ) over the Paris region, hereafter referred to as IDF1. For this run, we consider tabulated gas-particle aqueous equilibria for inorganic species, and chemically active aerosols. In this frame, the secondary organic aerosol

(SOA) is represented by six hydrophilic species, three hydrophobic species which are distinguished by their saturated vapor pressure and two substitution compounds representing the oxidation products of isoprene. Advection of all species is simulated using the Van Leer scheme, and we considered 8 vertical layers from ground level (997mbar) to 500 mbar. Further details on the processes simulated and the physical and chemical schemes available in the CHIMERE model can be found in (Menut et al., 2013) and references therein. As for meteorological inputs, we used simulations of the WRF model (https://www.mmm.ucar.edu/weatherresearch-and-forecasting-model) performed on 3 nested grids that are broader than the CHIMERE simulation domains in order to constrain the pollutant fluxes at the limits of the domains. The European domain is forced by NCEP meteorological fields (http://www.ncep.noaa.gov), and by analyzes of model outputs at a resolution of 1° every 6 hours. The WRF urban canopy module (UCM) was not activated due to the lack of descriptive data on urban coverage in the simulated areas. To compensate, we activated on all simulations the option of urban parametrization of CHIMERE which allows to correct the turbulence, and consequently the urban boundary layer height, over an anthropized area. This correction reduces wind speed at ground level by 20% and increases the sensitive heat fluxes at the surface. Finally, the anthropogenic emissions used in CHIMERE for the Paris region include the OLYMPUS emission data, supplemented for the airborne, industrial, agricultural and waste-related emissions by the regional kilometric inventory from the air quality monitoring network AIRPARIF. In the end, in the Paris region, the share of emissions created by Olympus represents 70% of the total nitrogen dioxide emissions and respectively 50% and 65% of the total mass of  $PM_{10}$ and PM2.5 emissions.



Fig. 2. Nested simulation domains and their horizontal resolution (on the left side), and urban versus forested and agricultural land use in the Paris region (on the right side).

## 3. CTM result analysis and processing

#### 3.1 Analysis of CHIMERE output concentration fields

The pollutant concentration data produced by CHIMERE are illustrated in Fig. 3 for NO<sub>2</sub>, in the form of a map of mean annual values (a) and along 3 time series (b) showing the distribution of the simulated values for each hour of the day in the 3 main areas of Île-de-France (the city center, the inner suburbs and the outer suburbs). The distribution of the hourly values, represented by a boxplot, reflects the temporal variability throughout the year, as well as the spatial variability inside each zone. This figure shows a satisfactory restitution of the well-known regional structure of the pollutant concentration fields. In particular, a 5-fold gradient is observed between the outer ring and the city center, where the levels reach  $30\mu g/m^3$  as an annual average, at the kilometric scale. This gradient is also observed in the model outputs for PM<sub>10</sub> (from 4 to  $20\mu g/m^3$ ) and to a lower extent for PM<sub>2.5</sub> (from 5 to  $13\mu g/m^3$ ). Thus, the fields have a concentric circle structure that underlines the centric organization of the modeled agglomeration. The dispersion of the values is also stronger in the dense zone, indicating greater spatial and/or temporal variability of the concentrations, characteristic of the proximity to emission sources.



Fig. 3. Spatial and temporal distribution of NO<sub>2</sub> concentrations modeled for the year 2009 with the OLYMPUS - CHIMERE platform. All concentrations are given in µg/m<sup>2</sup>. Upper map shows the annual average value. Lower time series show the average diurnal evolutions (hourly boxplots) by zone.

#### 3.2 Restitution of local concentration gradients

The CHIMERE concentration fields have been compared with measurements obtained at urban background and road traffic sites belonging to the AIRPARIF air quality monitoring network, both for nitrogen dioxide (19 / 7 sites respectively) and PM<sub>10</sub> (7 / 6 sites respectively). The results are illustrated in Fig.4. The upper left graph in Fig.4 underlines the relevance of the CHIMERE outputs in the assessment of NO<sub>2</sub> background pollution at all urban and peri-urban background sites, although we note that the model slightly underestimates the annual averaged observed values by 20-30%. Indeed, despite a remarkable capture of NO<sub>2</sub> temporal variations, CHIMERE simulates an annual average concentration of 23.4µg/m<sup>3</sup> at this grid point, while local measurements report 34.4µg/m<sup>3</sup>. This bias results from both a weaker baseline and an underestimation of most peak values in the model. One possible cause for this phenomenon may be the immediate vertical diffusion of pollutants in the first layer of the model, which can be excessive depending on the height of the meshes. At sites near traffic, despite much higher simulated values, the average and maximum levels of primary pollutants seriously underestimate the measurements. As mentioned in the introduction, this phenomenon is commonly reported for Eulerian model outputs. This results from the fact that the concentrations in the model are calculated at the scale of a mesh, from pollutant emissions which are averaged over this surface (here 1km²), while measurements made close to the circulation fully capture the variability of sources located only a few meters from them. It has been shown that such measurements have a representativeness area oriented along the road axis and which is at most 1 to 40m wide (Rodriguez et al., 2019). This is why it is now accepted that these two types of data are very poorly comparable quantitatively. However, it should be noted that qualitatively, the hourly evolution of NO2 concentrations under the effect of sources and daily atmospheric dynamics, is still correctly reproduced by CHIMERE. Regarding PM<sub>10</sub>, the concentrations are substantially underestimated compared to the measurements, whatever the type of site. We must conclude that beyond the above-mentioned phenomena, particle emissions in urban areas are undersized (lack of representation of organic compounds with moderate volatility for example, but also poor appreciation of the intensity of combustion sources). In all 4 cases, we clearly observe an underestimation of the baseline of the measured concentration, in combination with a too small amplitude in the variability of the simulated signal at the hourly scale - that is to say in the restitution of concentration peaks along the day.



Fig. 4. Comparison of the CHIMERE kilometric outputs (grey line) and the AIRPARIF monitoring network measurements (red line) for NO<sub>2</sub> (left column) and PM<sub>10</sub> (right column) in  $\mu$ g/m<sup>3</sup>, at peri-urban background sites (blue sticker, upper graphs) and at the traffic site located along the A1 highway (pink stocker, lower graphs), from October 19 to November 30, 2009. The lines in the upper left graph show the measured versus modeled mean annual concentration for this site.



Fig. 5. Organization chart of the downscaling method and illustration of the results obtained at the measurement site along the A1 Highway

		NO <sub>2</sub>					PM10				
Sites		Measured annual mean value (µg/m <sup>3</sup> )	Simulated annual mean value (µg/m <sup>3</sup> )	RMSE	Corrected annual mean value (µg/m <sup>3</sup> )	Reduction in RMSE	Measured annual mean value (µg/m3)	Simulated annual mean value (µg/m3)	RMSE	Corrected annual mean value (µg/m3)	Reduction in RMSE
	Issy	36,2	22,6	21,7	31,0	-21%	26,8	8,4	23,4	25,1	-37%
Urban background	La Défense	35,8	26,1	18,6	35,6	-19%	29,0	9,6	24,1	28,4	-39%
	Paris 1er	38,0	32,0	20,1	43,9	-10%	29,1	10,9	23,4	32,3	-31%
	Auteuil	113,2	27,1	94,0	100,8	-49%	49,6	8,9	45,9	42,9	-49%
Proximity to traffic	Basch	96,4	27,5	75,8	86,2	-48%	44,7	9,7	39,3	39	-48%
to duffe	Elysées	66,1	31,1	44,6	60,8	-35%	44,1	10,4	39,3	38,6	-45%

 Table 1. CHIMERE scores at representative urban background and traffic sites, for both NO2 and PM10 measurements. All data are from the learning subset of stations. The light grey columns show the statistics for the initial CHIMERE run, while the dark grey columns show the corrected run scores and the reduction of RMSE.

To deal with the insufficient representation of fine-scale phenomena in chemistry-transport models, we opted for a statistical downscaling approach which creates, downstream of the CTM and from its data, new information at the subgrid scale (Makni et al., 2019). The primary goal is to better represent two typical urban environments: urban background noise and the variety of exposures to road traffic on the streets. Figure 5 proposes several illustrations highlighting the successive steps of the downscaling methodology, which relies on two main stages. First, we carried out a learning phase centered on the systematic analysis and correction of the model error, using a subset of the AIRPARIF measurement sites available to us: half of the urban background sites, and n-2 traffic sites (due to their low number, we have chosen to prioritize the learning process), all randomly selected. To calculate the corrected signal, we started from our previous findings: the corrected value must propose both an enhancement of the background level (coefficient  $\alpha$  \* estimate of the urban background level) and an amplification of the hourly amplitude of the signal (coefficient  $\beta$  \* simulated hourly concentration). The running average over the 8 hours prior to the time step under consideration proved to be a good estimate of the urban background level. Thus, the corrected signal is calculated as the following linear combination

## $C_{CORR} = \alpha \overline{C_{CHIMERE 8h}} + \beta C_{CHIMERE}$

The alpha and beta values are obtained, at each site and for each targeted pollutant, by multiple regression between simulated concentrations (averaged and hourly) on the one hand and the concentrations measured at the learning point on the other hand, over the whole of 2009. The fitting criterion is the reduction of the root mean square error. As an example, for NO2, we obtained respective values of 1.9 and 1.6 for  $\alpha$  and  $\beta$  at the traffic site located Porte d'Auteuil (Paris ring road), and 0.7 and 0.5 for the urban site of Aubervilliers. It is important to note that we had to progressively increase the correction for a range of moderate (> 5 m/s) to strong (10 m/s) winds. Indeed, it appeared that the model error was larger in such situations. One possible reason is that the CTM does not take into account the geometry of buildings that act partially as a barrier to pollutant advection during moderately windy events. We applied an additional correction, represented by the factor  $(1 + \gamma)$  shown in the equation of Figure 5. The value of  $\gamma$ , which is unique for all sites and pollutants, follows a bell curve in order to be maximum at 8m/s and to tend towards 0 outside the wind range mentioned above. The  $\gamma$  value was set at 1 after statistical adjustment. With the  $\alpha$ ,  $\beta$  and  $\gamma$  coefficients thus obtained, we were able to provide locally corrected outputs at each site and for each pollutant. As can be seen in Fig. 5, the correction of CHIMERE outputs allows to obtain a background level in better agreement with the measurement, and to better reproduce the high values observed during the day, or during small pollution events. Table 1 presents the scores of the model before / after correction, on a set of learning sites representative of the obtained results. They show that the corrected evaluation of the average annual concentrations in each micro-environment is much more faithful to the observations. Furthermore, the root mean square error is significantly reduced, by almost a factor of 2 in proximity to traffic, and by 20 to 40% in urban areas. The lower RMSE reduction score for  $NO_2$  is linked to the fact that the signal is very noisy and that a slight time shift between model and measurements generates a random error which is not reducible by this approach. It should be noted that the correction only affects the intensity and amplitude of the signal, but that the temporal variability of the concentrations - quite well reproduced overall - is brought by CHIMERE outputs.

In a second step, the  $\alpha + \beta$  coefficients were analyzed to see if systematic patterns and behaviors could be found, with the aim of predicting their value in all urban and road environments, outside of learning points. The following conclusions were drawn:

- a) The ratio between the two coefficients is not very variable in the same environment (either urban background or traffic) and for a single pollutant. This illustrates the fact that the structure of the model error does not vary site by site but relies on the same drivers in a given environment. Thus, we simplified this result by using a unique total coefficient  $\alpha + \beta$  with a fixed speciation.
- b) For urban background sites, the total coefficient value showed little variability from site to site. It was fixed at the mean value of all sites, that is 1.3 for NO<sub>2</sub> and 2.8 for PM<sub>10</sub>.
- c) For traffic sites we were able to establish a robust correlation between the amplitude of the correction needed  $(\alpha + \beta)$  and the amplitude of the error due to the averaging of traffic emissions in each CTM mesh (1x1km<sup>2</sup>). The magnitude of this error called "averaging effect" is equal to the ratio between local traffic and total mesh traffic. In highly urbanized areas, this correlation did not appear to be disturbed by other emission effects. It should be mentioned that the built environment (open *versus* confined urban spaces) was also considered in this approach, but no proxy showed any correlation with the coefficients  $\alpha$  and  $\beta$ .

Using the traffic flows simulated by OLYMPUS on each road section of the domain, we were able to calculate the "averaging effect" on all roads in the urban domain and predict the value of  $\alpha + \beta$  at these locations. For urban background areas, we used the value of  $\alpha + \beta$  agreed upon earlier in b). We then applied these correction coefficients for the NO<sub>2</sub> and PM<sub>10</sub> concentration fields for the entire city, divided into "urban background" or "traffic proximity" areas. The validation of this approach was carried out by applying these coefficients to the AIRPARIF sites not used for the learning step. The results are illustrated in Fig. 6 and Fig.7 for stations of both types and validate our statistical approach. Indeed, they show that it offers a real opportunity to get closer to the NO<sub>2</sub> and PM<sub>10</sub> concentrations actually measured in the city, both quantitatively and over time variability.



Fig. 6. Boxplots illustrating the central parameters as well as the distribution of the NO<sub>2</sub> (top) and PM<sub>10</sub> (bottom) concentration values for the whole year 2009 at different validation sites. The AIRPARIF measurement data are shown in red, the original CHIMERE output in grey, and the corrected modeled values in green. The two sites on the left are located in (peri-)urban background areas, and the site on the right (national road 2 - RN2) is located in close proximity to road traffic.



**Fig. 7.** Comparison of the CHIMERE (grey line) and CHIMERE corrected (green line) outputs with the measurements of AIRPARIF at the traffic site located along the National Road 2 (RN2). For NO2 (upper panel), the period shown is that from November 17 to December 4<sup>th</sup>. For PM<sub>10</sub> (bottom panel), the period presented extends from November 1<sup>st</sup> to December 4<sup>th</sup>, 2009



Fig. 8. On the left map, NO<sub>2</sub> concentrations simulated by CHIMERE at the kilometric scale on January 2, 2009 for the 6:00-7:00 AM timestep (UT). On the right map, corrected CHIMERE outputs

The maps resulting from the application of this method to the entire agglomeration is presented in Fig. 8 for illustration: it shows concentration gradients that are more consistent with current knowledge on the inhomogeneity of air quality in a city. Given the extent of the corrections made to the model, these result challenge the modeling approaches that need to be set-up in the context of exposure evaluation and related decision support.

#### 3.3 Implementing a dynamic approach for exposure assessment

This section discuss the importance of implementing dynamic exposure calculation based on refined concentration data. It proposes to compare two different estimates of the exposure of an individual, provided on the one hand by the standard version of CHIMERE at the place of residence, and on the other hand by the corrected concentrations from OLYMPUS-CHIMERE-Downscaling and taking into account the mobility of this individual during the day. To do this, we used in the first case the 24-hourly data simulated at the grid point corresponding to the residence of an agent of the OLYMPUS synthetic population. In the second case, we used the daily mobility of this agent, which has been statistically estimated by the OLYMPUS tool from regional household surveys, to consider his presence in different urban microenvironments along the day. We chose to work on an individual living in the suburbs of Paris and making a daily return trip from home to work in a private vehicle (Fig. 9).

The distribution of NO<sub>2</sub> concentrations simulated by CHIMERE on the kilometric mesh corresponding to the dwelling of the selected individual, and during a typical day, is shown in green in the histogram of Fig. 10. It symbolizes the results of a "historical" approach to exposure. With regard to the dynamic approach using the fine-scale correction, we calculated the temporal distribution of the exposure of this same individual by taking into account 2 daily trips by car (pendulum mobility), and by weighting the concentrations along these trips by the time spent on each road axis. The box-plot on the left side of Fig. 10 illustrates the importance of using downscaling data, and not kilometric resolution data, in order to restitute the proximity between the agent and the traffic emissions in the vehicle. For this dynamic exposure calculation process, we considered that the individual - in his home and at his workplace - is exposed to an atmosphere which composition is that of the urban or peri-urban background where he is located. This is an approximation, which tends to consider that the indoor environments are perfectly ventilated and homogeneous with the outside atmosphere. This is realistic to a certain extent since it has been shown a very efficient transfer of external pollutants to the interior of buildings (Kirchner et al. 2001). However, it should be noted that this approximation may as well underestimate or overestimate the indoor concentrations, depending on the agent's activities. Indeed, household chores, cooking and continuous emissions of volatile compounds by living materials can significantly affect indoor air composition. This is why it is critical, in the long term, to implement a module for indoor atmosphere allocation, including exchanges with surrounding outside air. Recent works make it possible to envisage such a functionality (ANSES, 2019). In the end, the histogram shown in red in Fig. 10 represents the distribution of the exposure obtained using this innovative OLYMPUS-CHIMERE + downscaling and mobility approach.







Fig. 10. Comparison of the temporal distribution of the exposure of an individual to NO, calculated by a static approach that does not take into account the fine-scale urban concentration gradients (green series) *versus* an approach including the restitution of proximity to the traffic lanes and the daily mobility of the individual (red series).

The data shown in this last figure suggest that pollutant exposure in metropolitan areas may be much more heterogeneous than previously thought. In our case, indoor environments have not been specifically simulated, and mobility is reduced to commuting. Despite the low temporal representativeness of these events, the road micro-environments are likely to present strong break-up concentrations with the urban or peri-urban background atmosphere simulated at the CTM scale. It is legitimate to ask what, in this case, is the danger of chronic exposure to pollution? The annual average exposure - regardless of the data that compose it, or the recurring frequentation of highly polluted environments? Due to lack of input data, cohort approaches - and more broadly epidemiological studies, have not really addressed this issue. However, they have already been pointing out the populations living near the most frequented roads, as individuals undergoing strong environmental inequality.

The divergences observed between the approaches, in the distribution of the exposure values, therefore clearly conclude that there is a need to rethink our exposure assessment practices, in order to integrate a greater part of the urban reality - and the practices of the individuals - in the calculations. Such an approach would make it possible to discuss efficient ways of reducing population exposure to air pollutants.

#### 4. Conclusions

In this work, we have implemented a new platform for modeling air quality and the exposure of individuals, on the Greater Paris area, for the year 2009. The objective of this work was to evaluate the impact of a finer representation of air composition in dense urban areas – and especially near the traffic routes, but also to assess the potential contribution of the mobility of individuals, in the calculation of their exposure. The simulation works were carried out with the OLYMPUS tool, for the generation of an activity database centered on the individual and for the calculation of the associated atmospheric pollutant emissions. The emission dataset produced by OLYMPUS on this area has been validated in a previous study. The results of the air quality simulation conducted by CHIMERE, using the OLYMPUS outputs, showed a very good understanding of the pollution gradients observed at the scale of the metropolis. However, the physical processes on which this model is based, as well as the principle of a kilometric discretization, both limit its performances when reaching the street scale. We have shown that it is then critical to rework data from CTMs if we wish to take into account the large concentration gradients (up to a factor of 5) that prevail within the dense city. We chose to carry out a statistical correction of the CTM outputs that was conducted over a restricted set of learning sites, and that can be extended to any traffic or urban background area of the city, based on the local importance of traffic versus total traffic in the model mesh. The implementation of this approach allowed to quantify the gain in realism of the simulated data, compared to measurements made along the road axes.

Lastly, we addressed the question of exposure evaluation, by showing that standard approaches highly underestimate the values calculated using refined air quality data and daily trip of individuals in the city. For this purpose, we estimated the exposure of a given individual in Greater Paris, first using the CHIMERE concentration data at its place of residence, and then using a dynamic approach integrating the agent mobility and the statistically refined CHIMERE concentrations along its daily trips. This individual was randomly selected from the synthetic population created by OLYMPUS. Our results show that this mobility-based approach produces a much more heterogeneous set of exposure values than in the classical approach, with particularly high values during the trips. These high values do not significantly alter the average daily exposure (39  $\mu$ g/m<sup>3</sup> versus 28 $\mu$ g/m<sup>3</sup> on the selected day). However, these results raise the question of whether repeated (but short) stays in highly polluted microenvironments can be the drivers of the health effects observed in the cohorts related to cities with high traffic. Beyond this problem, the use of a dynamic exposure calculation avoids the bias related to the place of residence of individuals. Indeed, an inhabitant of the peripheral suburbs who is coming daily by car in the dense zone will undergo a much stronger exposure than if he carried out all of his activities close to his home. Thus, this approach opens the way towards new diagnoses about the link between urban form, location of jobs, transport networks, socio-professional practices and environmental inequalities. Finally, it is essential to include in these works a reflection on the impact that the refinement of exposure will have on the evaluation of the health effects linked to pollution. Indeed, taking into account a new range of exposure values for urban areas raises the question of the validity and use of dose-response curves. Today, with the rise of societal pressure on the control of urban air quality, health and environment research must open up to multidisciplinary issues and increase its level of technicality, in order to produce decision support that is relevant and adapted to the challenges of urban sustainability. In this frame, an innovating and interdisciplinary collaboration is expected between air quality researchers and epidemiologists around a more robust assessment of exposure to pollutants in urban areas and the associated health impacts.

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