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Effect of vehicle fleet composition and mobility on outdoor population exposure: A street resolution analysis in Paris

Lya Lugon^{a,b,*,1}, Youngseob Kim^a, Jérémy Vigneron^c, Olivier Chrétien^b, Michel André^d, Jean-Marc André^e, Sophie Moukhtar^{c,2}, Matteo Redaelli^f, Karine Sartelet^a

^a CERE, École des Ponts ParisTech, EDF R&D, 77455 Marne la Vallée, France

^b Paris Department of Green Spaces and Environment, 75013, Paris, France

^c AIRPARIF, 75004, Paris, France

^d Université Gustave Eiffel - IFSTTAR, Bron, 69675, France

^e Citepa, 75010, Paris, France

^f ANSES, 94701, Maisons-Alfort, France

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ABSTRACT

High concentrations of health-related pollutants are observed in streets, inducing high outdoor population exposure. The impact of reducing traffic emissions on urban air quality is often quantified using regional-scale chemical-transport models (CTMs), which are representative of background concentrations but fail to represent the high street concentrations. Local-scale models may be more suitable to represent them, but their use for outdoor exposure assessments is still limited, because they often neglect secondary species, use simplified methods to estimate background concentrations, and omit/underestimate non-exhaust emissions. Here the impact of realistic and ambitious tendencies of vehicle fleet renewal and mobility on outdoor population exposure is investigated using multi-scale simulations. They are capable of representing secondary species in urban background and streets (including the formation of secondary organic and inorganic aerosols), and use non-exhaust emissions determined with a recently revisited approach. The 10-year vehicle fleet renewal induces large decreases in population exposure to NO₂, black carbon, PM₁₀, PM_{2.5}, and organics. These decreases are stronger than estimated using CTMs. Favouring recent diesel, petrol, or electric vehicles, as proposed here, induces similar decreases of outdoor population exposure to PM_{2.5}. Promoting electric vehicles induces the highest decrease of exposure to NO₂. Home-office practice enhances the decrease of pollutant concentrations, but it is less effective than vehicle fleet renewal. However, much more ambitious reductions of pollutant emissions are still needed to respect air-quality guidelines.

1. Introduction

Atmospheric pollution is one of the most important risks to human health, causing about 7 million premature deaths worldwide (WHO, 2018). Cities are often subject to high concentrations of nitrogen dioxide (NO₂) and particles of diameter lower than 2.5 μm (PM_{2.5}) (Zhao et al., 2020; Shi et al., 2020; Ramacher and Karl, 2020), whose mass concentrations are regulated. There is evidence that NO₂ can deeply penetrate in the lung, inducing respiratory diseases. Different health impacts are related to high NO₂ concentrations, ranging from coughing, wheezing, but also asthma, dyspnoea, bronchospasm, and even pulmonary oedema in extreme cases (Kowalska et al., 2020; Cisneros et al., 2021; Manisalidis et al., 2020; Kowalska et al., 2020). The evidence

of adverse health effects of particles encompasses a broad spectrum of respiratory and cardiovascular outcomes, and there is emerging evidence related to neonatal and neurological outcomes (Dedoussi et al., 2020; Fischer et al., 2020; Andreão et al., 2020; Niu et al., 2021). These effects depend on the particle size and chemical composition (Dedoussi et al., 2020; Fischer et al., 2020; Fang et al., 2016; Crobeddu et al., 2017; Cervellati et al., 2020; Kwon et al., 2020). Notably black carbon (BC) and organic compounds of anthropogenic origins (PM_{org,anth}) may strongly affect human health (Mudway et al., 2020; Ali et al., 2020; Quang et al., 2021; Pant and Harrison, 2013; Daellenbach et al., 2020; Farzad et al., 2020; Lequy et al., 2021). BC is emitted by road traffic and combustion processes, while PM_{org,anth} is both emitted

* Correspondence to: CERE, École des Ponts ParisTech, EDF R&D, 77455 Marne la Vallée, France.
E-mail address: lya.lugon@enpc.fr (L. Lugon).

¹ Now at: Max Planck Institute for Meteorology, 20146, Hamburg, Germany.

² Now at: Citepa, 75010, Paris, France.

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and partly formed in the atmosphere (secondary compounds). Recent studies pointed out that $PM_{org,anth}$ may be more dangerous to human health than organic compounds of biogenic origins (Daellenbach et al., 2020; Farzad et al., 2020; Lequy et al., 2021; Nault et al., 2020).

Several modelling studies estimated the impact of road traffic on atmospheric pollution, using hypothetical scenarios of vehicle fleet composition (Roustan et al., 2011; Andre et al., 2020; Samuelsen et al.; Yang et al., 2020; Zhang et al., 2020; Minet et al., 2020), or mobility scenarios during the COVID-19 lockdown (Menut et al., 2020; Huang et al., 2021). They point out that a decrease of traffic emissions strongly reduces NO_2 and BC concentrations, but has a lower impact on $PM_{2.5}$. Typically in the Parisian region, studies show that a reduction of NO_2 traffic emissions by around 50% leads to a decrease of NO_2 concentrations by about 25% to 40% (Roustan et al., 2011; Andre et al., 2020). For $PM_{2.5}$, a sharp decrease in traffic emissions reaching 80% leads to a limited decrease of $PM_{2.5}$ urban concentrations (by about 15%) (Roustan et al., 2011). This limited decrease can be explained by the importance of long-range transport of $PM_{2.5}$, involving distant sources (e.g. industries, agriculture) and other local sources (e.g. residential activities, cooking). Furthermore, as the fleet is renewed, promoting recent petrol vehicles seems to be more efficient than diesel at limiting NO_2 , and to a lesser extent $PM_{2.5}$ concentrations (Andre et al., 2020). These studies use regional-scale chemical-transport models (CTMs) (Sartelet et al., 2007; Zhang et al., 2010; Menut et al., 2013; Byun and Ching, 1999; Mensink et al., 2001), which are able to represent the impact on background concentrations, including the formation of secondary compounds. However, they fail to represent the high concentrations observed in streets, to which populations are exposed.

These modelled background concentrations are also often used to estimate population exposure and health effects (Kelly et al., 2011; Ostro et al., 2015), even though they cannot represent the real population exposure taking into account commuting and indoor environments. Although outdoor exposure may represent only a small fraction of the population exposure depending on the seasons, it needs to be carefully assessed as it affects many environments to which people are exposed (Soares et al., 2014; Ramacher and Karl, 2020). Background concentrations do not represent the high concentrations observed in streets, which are those to which populations are exposed. Notably in large megacities such as Paris city, high concentrations in streets are of high concern for air quality and urban mobility policies (Pascal et al., 2013). A wide range of local-scale models exist to represent the street concentrations and sometimes to model population exposure (Yamartino and Wiegand, 1986; Eerens et al., 1993; Berkowicz, 2000; Cimorelli et al., 2005; Soulhac et al., 2011; Stocker et al., 2012; Briant and Seigneur, 2013; Sharma et al., 2013; Soulhac et al., 2016; Kim et al., 2018).

Notably in large megacities, such as Paris city, high concentrations of atmospheric pollutants are observed in streets, becoming a high concern for air quality and urban mobility policies (Pascal et al., 2013). A wide range of local-scale models exist to represent the street concentrations (Yamartino and Wiegand, 1986; Eerens et al., 1993; Berkowicz, 2000; Cimorelli et al., 2005; Soulhac et al., 2011; Stocker et al., 2012; Briant and Seigneur, 2013; Sharma et al., 2013; Soulhac et al., 2016; Kim et al., 2018). Some of them have been used in different cities to show that vehicle fleet renewal and new urban mobility policies may strongly reduce the population exposure to NO_x (Tezel-Oguz et al., 2020), PM_{10} (Maesano et al., 2020), NO_2 (Minet et al., 2020; Host et al., 2020), and BC, with lower reductions of population exposure to $PM_{2.5}$ (Minet et al., 2020). These studies are still scarce, as local-scale models have important limitations: they often adopt simplifications to determine background concentrations, they ignore secondary particle formation, or adopt a non-realistic stationary hypothesis to calculate reactive pollutant dispersion, or even neglect or underestimate non-exhaust emissions. Some models bridge the gap between regional and local scales by including street-canyon parametrizations in urban-scale models, e.g. EPISODE-CityChem v1.2 (Karl et al.,

2019), UBM/OSPM (Ketzel et al., 2021), or by using downscaling technique, e.g. uEMEP_v5 (Denby et al., 2020). The recent street-network Model of Urban Network Intersecting Canyons and Highways (MUNICH) (Kim et al., 2018; Lugon et al., 2021) overcomes these limitations. MUNICH integrates a nonstationary approach to split transport and chemistry, which improve the representation of reactive compounds (Lugon et al., 2020a); and presents an innovative approach to estimate particle resuspension, strictly respecting the mass balance over the street surface (Lugon et al., 2020b). However, the biggest difference of MUNICH compared to the others mentioned models is linked to the Eulerian approach used to discretize street segments, that allows the integration of a robust chemical module (SSH-aerosol (Sartelet et al., 2020)) capable of representing gas-phase chemistry, particle dynamics, as well as SOA and SIA formation in the streets [50]. Furthermore, MUNICH enables a multi-scale modelling approach with a one-way coupling with the 3D Eulerian CTM Polair3D (Sartelet et al., 2007) (the background concentrations calculated by Polair3D are inputs to calculate concentrations in streets with MUNICH). Both Polair3D and MUNICH use the same chemical module SSH-aerosol in the regional and local scales, assuring a consistent representation of chemistry across the scales.

Building-up on the recent capabilities of multi-scale modelling, this study investigates the influence of vehicle fleet composition and urban mobility on the residential exposure of the Parisian population to outdoor NO_2 , $PM_{2.5}$, PM_{10} , BC and $PM_{org,anth}$. Different traffic emission scenarios are considered to represent the vehicle fleet compositions and urban mobility hypotheses. They model the evolution of the 2014 Parisian vehicle fleet composition over ten years, promoting either recent diesel (2-BAU scenario), or petrol (3-PET scenario) or electric vehicles (4-ELEC scenario). These emission scenarios are detailed in Section 2 and in Andre et al. (2020), which previously simulated the impacts of 2-BAU and 3-PET scenarios at the regional scale over France and Île-de-France region. Compared to Andre et al. (2020), non-exhaust emissions are revisited based on recent estimations (Lugon et al., 2020b). Also, an additional emission scenario is added, based on 3-PET, to represent the influence of home-office practice (5-PET_{ho}) increasingly applied in Paris since the beginning of the COVID-19 epidemic. To estimate the impact of these scenarios on the population exposure, multi-scale modelling is used here to simulate the concentrations of multiple compounds. As mentioned, the urban background concentrations (regional scale) are simulated using the CTM Polair3D (Sartelet et al., 2007). It is coupled to MUNICH (Lugon et al., 2021) to calculate the local-scale concentrations of the main 3700 streets of the Parisian street-network with a consistent multi-scale system.

2. Traffic emission scenarios

Pollutant emissions are computed from four realistic and ambitious scenarios representing different tendencies of traffic fleet renewal over ten years and of urban mobility over Paris. The reference simulation corresponds to the year 2014 (1-REF). Three scenarios concern the traffic fleet composition, promoting different technological options in vehicles engines. As detailed in Andre et al. (2020), diesel is promoted in the 2-BAU scenario, essence in the 3-PET scenario and electric vehicles in the 4-ELEC scenario. An additional scenario is proposed here. It integrates in the 3-PET scenario the mobility changes induced by home-office practice (5-PET_{ho} scenario). Home-office practice is increasingly recommended in France, notably with the COVID-19 sanitary crisis. All scenarios use European regulatory emission standards (pre-Euro 2 to Euro 6) to estimate pollutant emissions from the different vehicle categories (light-duty vehicles (passenger cars and vans), heavy-duty vehicles (trucks and buses), and motorized two-wheelers). Fig. 1, from (Andre et al., 2020), illustrates the vehicle fleet composition of each scenario in terms of engine and emission standards for the Île-de-France region. In each scenario, the vehicle fleet is renewed and the main characteristics are summarized below:

- 1-REF: The reference simulation represents the 2014 emissions. It was calculated based on the 2014 vehicle fleet composition.
- 2-BAU: The “business-as-usual” scenario promotes recent diesel vehicles with particulate filters (Euro 5 and Euro 6 vehicles according to European regulatory emission standards). This scenario assumes almost the same balance between engine types (petrol, diesel and electric) and vehicle categories, as in 2014.
- 3-PET: This scenario promotes petrol rather than diesel light vehicles. It assumes a steady decline in diesel vehicle sales over ten years; from 60% to 5%, decreasing the percentage of diesel light-duty vehicles (passenger cars and vans) from 73% in the 1-REF situation, to 43% in 3-PET scenario. The other vehicle categories present the same regulatory standards and fuel engine as used in the 2-BAU scenario.
- 4-ELEC: This scenario promotes electric vehicles in all vehicle categories. As mentioned in Andre et al. (2020), 4-ELEC scenario considers electric vehicle sales reaching 40% of passenger cars and 60% of light-duty vehicles in 2025. The final percentage of electric vehicles in each vehicle category is 11% for light-duty vehicles, 2.3% for trucks, 15% for buses and 42% for motorized two-wheelers.
- 5-PET_{ho}: As the scenario 3-PET, this scenario promotes petrol light vehicles. However, urban mobility is modified to represent the reductions of mobility induced by home-office practice. Note that the objective in analysing this scenario is to evaluate the benefits of home-office practice as a long-term practice generalized over the city to improve air quality. For this, mobility data from the COVID 19 crisis are used, and the following hypothesis are considered: (i) mobility reductions do not affect the fleet composition; (ii) mobility reductions only affect passenger cars, and not the other vehicle types; (iii) home-office practice is only during the weekdays, not at the week-ends. The mobility reduction is expressed by a reduction in passenger car traffic emissions (f_{ho}). This reduction factor f_{ho} is a function of the number of home-office days per week (N_{ho}), the number of workdays per week (N_w), the ratio of the Île-de-France population that adopts home-office practice (p_{ho}), the reduction displacement ratio between home-office days and workdays (d_{ho}), as shown in the equation below:

$$f_{ho} = 1.0 - \left(\frac{N_w - N_{ho}}{N_w} + \frac{N_{ho}}{N_w} \times [(1.0 - p_{ho} + p_{ho} \times d_{ho})] \right) \quad (1)$$

Three days are considered to be home-office days ($N_{ho} = 3$) out of five workdays per week ($N_w = 5$). According to a recent survey performed in the Île-de-France region (Pillouy et al., 2020), around 56% of workers adopted home-office as a current practice during the COVID-19 lockdown ($p_{ho} = 0.56$). The latest mobility report published by the Île-de-France mobility observatory (OMNIL) (OMNIL, 2019) indicates that 38% of the total kilometres travelled in the region are correlated to work-home displacements. As there is no work-home displacements during home-office days, the total vehicle displacements correspond then to 62% of the displacements of normal workdays ($d_{ho} = 0.62$). With all these considerations, home-office practice induces an average reduction of 13% of passenger car mobility compared to weekdays ($f_{ho} = 0.13$). Because traffic emissions and vehicle displacements are linearly related (emissions are calculated using emission factors (Franco et al., 2013), in $\mu\text{g} (\text{veh km})^{-1}$), as a first approach, this reduction ratio is applied to passenger car emissions.

As detailed in Andre et al. (2020), for the reference simulation, traffic emissions were calculated using hourly flow and speed data observed during 2012 in more than 40,000 street segments in the Île-de-France region. It includes the Paris City street-network with the main 3700 street segments, which are used in the local-scale simulations. Exhaust and evaporative emissions for carbon monoxide

Table 1

Traffic emissions over Paris city street-network with the 1-REF reference (in t yr^{-1}) and relative differences between traffic emissions from the different scenarios compared to 1-REF (in %).

	NH ₃	SO ₂	NO ₂	NMHC	PM ₁₀	PM _{2.5}	BC	PM _{org,anth}
1-REF [t yr^{-1}]	189.9	26.0	3,656.0	3,510.7	2,611.2	796.7	890.4	686.8
2-BAU [%]	-53	-38	-58	-51	-41	-54	-51	-37
3-PET [%]	-44	-36	-68	-51	-41	-60	-56	-35
4-ELEC [%]	-55	-45	-69	-66	-41	-59	-54	-37
5-PET _{ho} [%]	-49	-39	-70	-51	-44	-61	-58	-38

(CO), non-methane hydrocarbons (NMHC), nitrogen oxides (NO_x and NO₂), ammonia (NH₃), particle matter with diameters lower than 2.5 μm (PM_{2.5}) were computed by AIRPARIF using the Heaven platform with COPERT 4 emission factors adapted to different vehicle engines, emission standards and operational conditions. The Heaven platform estimates NO₂ emissions from NO_x emissions using NO₂/NO_x ratio from COPERT-IV. This ratio depends on the characteristics of the vehicle fleet (Tomassini, 2003). Particulate matter from exhaust emissions are assigned to PM₁, and assumed to be composed of organic matter and black carbon. Tyre, brake and road wear are responsible for all traffic emissions of particles of diameters higher than 2.5 μm . PM₁₀ non-exhaust emissions from brake and road wear are computed using COPERT 4 emission factors, with the chemical speciation of black carbon and organic matter proposed in the EMEP guidelines. PM₁₀ tyre wear emission factors are determined from the estimation of Lugon et al. (2020b) based on numerical simulations and the literature (Luhana et al., 2004; Boulter, 2005; Councell et al., 2004). The chemical speciation of tyre wear emissions is 25% of black carbon (the average BC mass weight fraction should be between 22%–30% (Thorpe and Harrison, 2008)), 36% of organic matter (as indicated in the EMEP guidelines (Ntziachristos and Boulter, 2016)), and the remaining 39% is classified as non-defined/dust. At emission, the PM_{2.5}/PM₁₀ ratios are the same as in the NORTRIP model (Soulhac et al., 2011), that is 4.5% for road wear, 10% for tyre wear and 40% for brake wear emissions.

The annual road traffic emissions in the Parisian street-network are displayed in Table 1 for the reference simulation (1-REF), as well as the relative difference of emissions between each scenario and the reference. All scenarios lead to significant decrease in traffic emissions of gas and particulate species. This decrease is due to the traffic fleet renewal. Regarding the influence of the traffic fleet (comparison of the 2-BAU, 3-PET and 4-ELEC scenarios), the most important decrease in gaseous emissions is observed in the 4-ELEC scenario, reaching an average decrease of 66% for NMHC and 69% for NO₂ emissions. Promoting recent petrol vehicles (3-PET scenario) rather than diesel vehicles (2-BAU scenario) results in a higher decrease of NO₂, PM₁₀, PM_{2.5} and BC emissions, but a lower decrease in NH₃ emissions, as NH₃ is mainly emitted by petrol vehicles. Significant differences of NMHC emissions are observed between the scenario 4-ELEC and the scenarios 3-PET and 2-BAU. NMHC is mainly emitted by petrol motorized two-wheelers: they correspond to 7.8% of total traffic, but are responsible for 49% of NMHC emissions in both the 3-PET and 2-BAU scenarios. However, 42% of motorized two-wheelers in the 4-ELEC scenario are electric vehicles, and 51% petrol vehicles, justifying the lowest NMHC emissions. Also, the second major source of NMHC from road traffic are evaporation emissions, which are around 70% higher in the 3-PET scenario than in 2-BAU and 4-ELEC scenarios. Even if the decrease of particle emission is quite similar in all scenarios, 3-PET presents the higher decrease. As shown in Fig. 1, the 3-PET scenario presents the highest percentage of recent light duty vehicles (92%, in contrast with 79% and 83% in the 2-BAU and 4-ELEC scenarios, respectively). Also, even if the 4-ELEC scenario promotes electric vehicles, it still assumes a relatively low percentage of electric light duty vehicles (11%), which limits the decrease of exhaust emissions.

The contribution of particulate exhaust and non-exhaust emissions in each traffic emission scenario is displayed in Table 2. For PM₁₀,

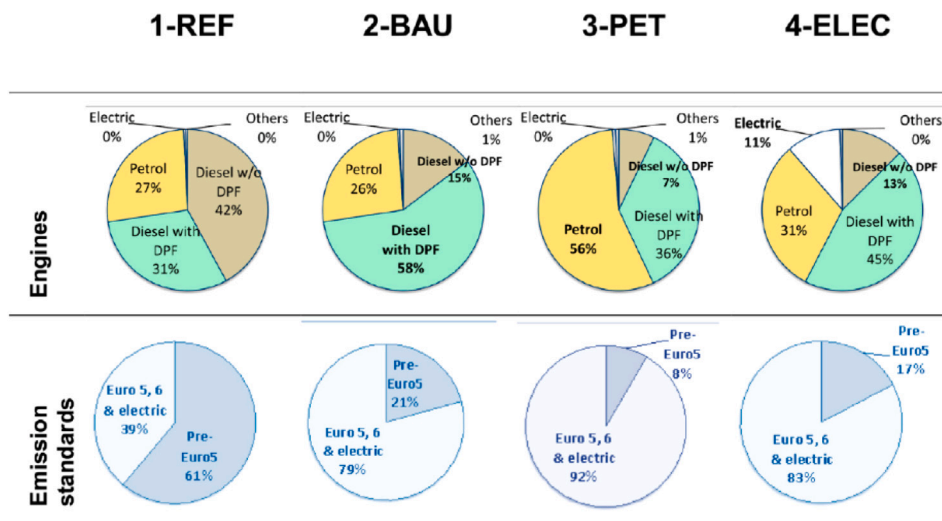


Fig. 1. Fleet composition of light-duty vehicles in the Île-de-France region (Andre et al., 2020).

Table 2

Contribution of exhaust and non-exhaust emissions to particulate traffic emissions in Parisian street-network for each traffic emission scenario. For each scenario and for each pollutant, the sum of exhaust and non-exhaust emissions is equal to 100%.

		PM ₁₀	PM _{2.5}	BC	PM _{org,anth}	DU
1-REF	Exhaust [%]	20	66	52	7	0
	Non-exhaust [%]	80	34	48	93	100
2-BAU	Exhaust [%]	12	52	37	7	0
	Non-exhaust [%]	88	48	63	93	100
3-PET	Exhaust [%]	9	43	28	6	0
	Non-exhaust [%]	91	57	72	94	100
4-ELEC	Exhaust [%]	10	45	32	4	0
	Non-exhaust [%]	90	55	68	96	100
5-PET _{ho}	Exhaust [%]	9	45	28	7	0
	Non-exhaust [%]	91	55	72	93	100

in the reference and in the scenarios, the contribution of non-exhaust emissions is higher than exhaust emissions: 80% in the reference and between 88% and 91% in the scenarios. This can be explained by the increasing technological improvements to limit particulate exhaust emissions of recent vehicles (from Euro 5 emission standards). It also explicits the importance to better understand and control non-exhaust emissions, still not well known in the literature and not controlled in the actual emission standards. Primary anthropic organic particulate matter is mostly emitted by non-exhaust emissions. However, it does not account for semi-volatile organic compounds emitted by exhaust emissions in the gas-phase. Previous simulations performed using MUNICH show their importance for organic particulate concentrations (Lugon et al., 2021). Note that due to the lack of chemical speciation of non-exhaust emissions, all emissions different from BC and organic matter are classified as dust (DU).

Note that the scenarios presented are consistent with the traffic fleet configuration observed nowadays in Paris. According to observations from the French Ministry of Ecological Transition (<https://www.statistiques.developpement-durable.gouv.fr/382-millions-de-voitures-en-circulation-en-france>):

1. A quite low increase of the total vehicle fleet flow is observed between 2014 and 2020 (4.5%)
2. Even if non-negligible differences exist regarding the vehicle technologies and Euro standards in the traffic fleet observed in Paris from 2014 to nowadays, quite similar fractions of diesel and petrol vehicles are observed in 2014 and 2020. The national fleet presented 63% of petrol-powered vehicles and 36% of

diesel-powered vehicles in 2014, and 59% of petrol-powered vehicles and 39% of diesel-powered vehicles in 2020.

3. The proportion of electric and hybrid particular vehicles fleet observed in Île-de-France region by the French Ministry of Ecological Transition in January 2021 is still low (around 0.3%), and far from the 11% adopted in the 4-ELEC scenario.

With the low fraction of electric vehicles in the current fleet, the scenarios presented here still seem ambitious. Therefore, the conclusions obtained, even with a reference situation in 2014, can still support coming actions to reduce concentrations in urban areas.

Even if non-negligible differences exist regarding the vehicle technologies and Euro standards in the traffic fleet observed in Paris in 2014 and nowadays, these scenarios remain relevant, as the total vehicle fleet flow is similar. Furthermore, with the low fraction of electric vehicles in the current fleet, the scenarios presented here still seem ambitious. The conclusions obtained, with a reference situation in 2014, can still support future actions to reduce concentrations in urban areas.

3. Air quality simulations

3.1. Modelling air quality in the urban background

The 3D Eulerian CTM Polair3D (Sartelet et al., 2007) is applied to estimate air quality over the Île-de-France region for the reference simulation and for the scenarios. Polair3D numerically solves the chemical-transport equation considering anthropogenic and biogenic emissions, physico-chemical transformations, advection, diffusion, and dry and wet deposition. Polair3D is coupled to the chemical module SSH-aerosol (Sartelet et al., 2020) to represent the formation of secondary gas and particles in the urban background. Simulations over Île-de-France with the different emission scenarios were performed over one year, with a spatial resolution of 2 km × 2 km and 14 vertical levels. Boundary and initial conditions are obtained from the one-way nesting simulations with Polair3D over Europe and metropolitan France presented in Andre et al. (2020). Only the reference and the scenarios 2-BAU and 3-PET were simulated over France. The France simulations of the reference and the 2-BAU scenarios respectively are used as boundary conditions for the reference and 2-BAU scenarios over Île-de-France, while the France simulation of the 3-PET scenario is used as boundary condition for the 3-PET, 4-ELEC and 5-PET_{ho} scenarios. Anthropogenic emissions in the Île-de-France simulations are calculated using the 2014 European Monitoring and Evaluation Program (EMEP) emission inventory in regions outside the Île-de-France limits, and the emission inventory provided by the air-quality agency of Paris

(AIRPARIF) inside the Île-de-France limits. Road traffic emissions are calculated based on the 2014 AIRPARIF traffic emission inventory, as described in Section 2. Anthropogenic emissions from sectors other than road traffic are calculated using the 2012 AIRPARIF emission inventory. Note that, as mentioned in Lugon et al. (2020a), differences between 2012 and 2015 emission inventories are low for sources other than traffic. Therefore, using the 2012 inventory for sources other than traffic should not significantly impact the results of this study. Total NO_x emissions are split adopting a ratio of 9.2% of NO_2 , and the rest considered as NO, as often used in modelling studies over Europe and European cities. Biogenic emissions are estimated with the Model of Emissions of Gases and Aerosols from Nature (MEGAN v2.04) (Guenther et al., 2012). Meteorological data are calculated using the Weather Research and Forecasting (WRF) model version 3.9.1.1, with a two-way nesting approach (Grell et al., 2005) and a nudging technique to incorporate observational data from the Paris Orly meteorological station. All details about the WRF simulations, including the physical options adopted are available in Lugon et al. (2020a). The parametrization proposed by Zhang et al. (2003) is used to calculate dry deposition velocities of gas-phase compounds, and below-cloud scavenging is computed according to Sportisse and Du Bois (2002). More information about the deposition schemes used are presented in Sartelet et al. (2007).

3.2. Modelling air quality in the streets

The Model of Urban Network of Intersecting Canyons and Highways (MUNICH) (Lugon et al., 2021) is employed to simulate air quality in the Parisian streets. MUNICH estimates the evolution and formation of primary and secondary gas and particles in streets, as it is coupled to the chemical module SSH-aerosol (Sartelet et al., 2020). MUNICH integrates the background concentrations calculated by the regional-scale model Polair3D (Sartelet et al., 2007), with a one-way coupling technique. The street concentrations are computed using a non-stationary approach, providing a stable representation of reactive species and a better representation of the NO_2/NO partitioning (Lugon et al., 2020a). At the local scale, simulations are performed in a street-network composed of the 3700 main streets of Paris city. Width, length and building height of each street are defined as described in Lugon et al. (2020a), using different databases and open areas as gardens and parks. Road traffic emissions are calculated in each street as described in the section on the traffic emission scenarios, based on the 2014 AIRPARIF traffic emission inventory. Meteorological data are the same as those used in the regional-scale simulations with Polair3D. Note that a consistent coupling between the regional and local-scale simulations is performed, as at both scales the same chemical module, meteorological data, and traffic emission hypothesis are adopted.

3.3. Data to model comparison: validation of the reference simulation

3.3.1. Air-quality stations and performance criteria

Several air-quality stations monitor regulated pollutant concentrations in the urban background and in the streets of Paris. PM_{10} , $\text{PM}_{2.5}$, NO_2 and total NO_x concentrations observed at air-quality stations compare well to those simulated in the urban background and in streets using the reference emission scenario (1-REF). Figure 1 in the Supplementary Material section illustrates the location of Paris city air-quality stations, differentiating the five urban stations with blue dots (PA04C, PA07, PA12, PA13 and PA18), and the nine traffic stations with red dots (AUT, BP_EST, ELYS, HAUS, OPERA, CELES, BASCH, SOULT and BONAP). Two distinct performance criteria are usually employed to evaluate simulations, as indicated by Hanna and Chang (2012) and Herring and Huq (2018). To reach the most strict criteria, the statistical indicators should be in the ranges $-0.3 < \text{FB} < 0.3$; $0.7 < \text{MG} < 1.3$; $\text{NMSE} < 3$; $\text{VG} < 1.6$; $\text{FAC2} \geq 0.5$; and $\text{NAD} < 0.3$. To reach the less strict criteria, accepted in urban areas considering

Table 3

Statistical indicators at air-quality stations located in the urban background (bg) and in the streets (st) (s and o represent the average simulated and observed concentrations in $\mu\text{g m}^{-3}$, respectively).

Compound	s	o	FB	MG	NMSE	VG	FAC2	NAD
<i>bg</i>								
PM_{10}	23.5	22.6	0.04	1.05	0.19	1.13	0.93	0.14
$\text{PM}_{2.5}$	19.4	15.3	0.24	1.32	0.28	1.22	0.88	0.19
NO_2	43.2	39.0	0.10	1.04	0.18	1.16	0.91	0.15
NO_x	63.0	57.9	0.07	1.00	0.30	1.20	0.88	0.18
<i>st</i>								
PM_{10}	35.64	33.10	0.08	1.06	0.20	1.17	0.90	0.16
$\text{PM}_{2.5}$	24.96	20.73	0.18	1.21	0.22	1.19	0.89	0.16
NO_2	73.00	67.62	0.10	1.10	0.30	1.24	0.87	0.17
NO_x	150.28	198.50	-0.21	0.82	0.47	1.76	0.68	0.24

the difficulties to simulate concentrations in these areas, the statistical indicators should be in the ranges $-0.67 < \text{FB} < 0.67$; $\text{NMSE} < 6$; $\text{FAC2} \geq 0.3$; and $\text{NAD} < 0.5$. All statistical indicators are defined in Appendix A.

3.3.2. Validation of the reference simulation (1-REF)

A good agreement is obtained between PM_{10} , $\text{PM}_{2.5}$, NO_2 and total NO_x concentrations simulated and observed at Parisian air-quality stations, for both the regional and local scales. Figs. 2 and 3 illustrate, respectively, the $\text{PM}_{2.5}$ and NO_2 temporal evolution of simulated and observed daily concentrations at two Parisian air-quality stations: a regional-scale (urban background) station (PA04C) on the left pane, and a local-scale (traffic) station (AUT) on the right panel. The concentrations measured at the urban background stations are compared to the regional-scale concentrations simulated by Polair3D, while the local-scale concentrations are compared to the street concentrations simulated by MUNICH. A good representation of fine particle concentrations over 2014 is obtained, even during the pollution peak observed in March 2014. An overestimation of concentrations is observed in winter, possibly caused by an overestimation of anthropogenic emissions, and/or an underestimation of the atmospheric boundary layer height.

Tables 3 summarizes the average statistical indicators obtained for PM_{10} , $\text{PM}_{2.5}$, NO_2 and total NO_x over 2014, at urban background stations and at traffic stations respectively. The performance criteria indicated for urban areas are largely met for all compounds at both spatial scales. The most strict performance criteria are almost met for all pollutants (with a limit value 1.32 instead of 1.3 for MG regarding $\text{PM}_{2.5}$ in the urban background, and 1.76 instead of 1.6 for VG regarding NO_x in the streets). Supplemental material section presents the statistical indicators obtained at each urban background and traffic station separately. Total NO_x concentrations are composed of NO_2 and NO, and they tend to be underestimated at traffic stations located in large streets and open areas, as BASCH. As mentioned in Lugon et al. (2020a), in these streets the short lifetime of NO leads to high uncertainties in NO dispersion, questioning the assumption of uniform concentrations in the streets adopted in MUNICH in these cases. For the regulated pollutants (PM_{10} , $\text{PM}_{2.5}$ and NO_2), the performance criteria accepted in urban areas are met at each urban background and traffic station. The most strict performance criteria are also met at the majority of stations. Also, quite similar $\text{PM}_{2.5}/\text{PM}_{10}$ ratios as those observed are simulated both at urban background stations (0.8 simulated and 0.7 observed) and in streets (0.7 simulated and 0.6 observed). This $\text{PM}_{2.5}/\text{PM}_{10}$ ratio is lower in the streets than in the background because of the larger influence of coarse particles from non-exhaust emissions.

4. Impact of the emission scenarios on pollutant concentrations

In all simulations (1-REF, 2-BAU, 3-PET, 4-ELEC and 5-PET_{ho}) concentrations are higher in streets than in the urban background, especially for species related to health impacts, such as $\text{PM}_{org,anth}$ and

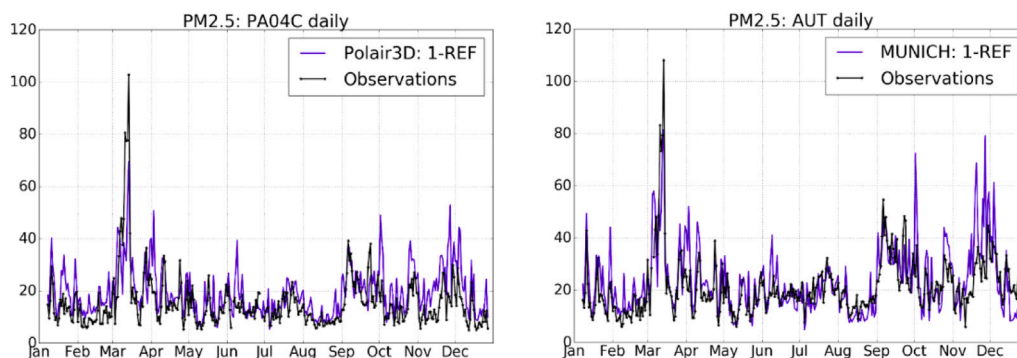


Fig. 2. Temporal evolution of PM_{2.5} daily concentrations simulated by Polair3D at the PA04C urban background station (left panel), and by MUNICH at the AUT traffic station (right panel).

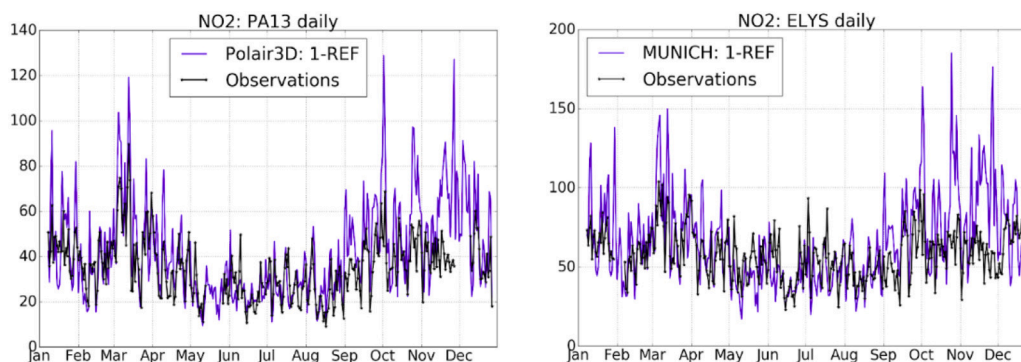


Fig. 3. Temporal evolution of NO₂ daily concentrations simulated by Polair3D at the PA13 urban background station (left panel), and by MUNICH at the ELYS traffic station (right panel).

Table 4

Average concentrations simulated with the 1-REF simulation (in µg m⁻³), and the average NMB (in %) between the scenarios and the 1-REF concentrations in streets (*st*) and in the urban background (*bg*).

	NO ₂		PM ₁₀		PM _{2.5}		BC		PM _{org}		PM _{org,anth}	
	<i>st</i>	<i>bg</i>	<i>st</i>	<i>bg</i>	<i>st</i>	<i>bg</i>	<i>st</i>	<i>bg</i>	<i>st</i>	<i>bg</i>	<i>st</i>	<i>bg</i>
1-REF [µg m ⁻³]	70.4	34.6	35.3	21.6	23.7	18.3	5.9	2.0	9.4	5.2	7.0	2.4
2-BAU [%]	-38	-33	-22	-11	-19	-10	-48	-40	-24	-11	-32	-20
3-PET [%]	-45	-38	-22	-11	-19	-10	-51	-44	-23	-10	-31	-19
4-ELEC [%]	-50	-39	-23	-12	-21	-10	-51	-43	-25	-11	-34	-21
5-PET _{ho} [%]	-47	-39	-25	-12	-21	-12	-54	-47	-27	-12	-37	-22

BC. Besides the higher concentrations observed in streets (where populations are exposed) than in the urban background, the air-quality benefits of reducing traffic emissions are also higher in streets. Table 4 shows the annual average concentrations in the streets (*st*) and in the urban background (*bg*) (first line), as well as the impact of each scenario on background and street concentrations, quantified by the average normalized mean bias (NMB, see Statistical indicators section). In the reference simulation (1-REF), the ratio between simulated concentrations in streets and those in the urban background ($r_{st,bg}$) are around 2 for NO₂, 1.6 for PM₁₀ and 1.3 for PM_{2.5}. Similar ratios between street and background concentrations were measured at Parisian air-quality stations over 2014 (1.7 for NO₂, 1.5 for PM₁₀ and 1.3 for PM_{2.5}). Large ratios were observed for BC and PM_{org} during an experimental campaign performed by AIRPARIF in 2009/201053 at two air-quality monitoring stations (AUT and PA04C): 4.9 for BC and 1.8 for PM_{org} (see Tables 1 and 2 in Supplemental Material section). Similar values of $r_{st,bg}$ are simulated with the 1-REF simulation: 4.3 for BC and 1.6 for PM_{org}. The ratio between simulated concentrations of PM_{org,anth}/PM_{org} are also higher in the streets (0.7) than in urban background (0.4), indicating the larger influence of anthropogenic traffic emissions in streets. In the urban background, other sources, such as biogenic, may strongly contribute to PM_{org}.

All the concentrations are reduced in the scenarios compared to the reference. The air-quality benefits of reducing traffic emissions are also higher in streets than in the urban background. The average decrease of PM₁₀, PM_{2.5}, PM_{org} and PM_{org,anth} concentrations in streets (from 19% to 37%) is about twice higher than in the urban background (from 10% to 22%). The impact of traffic scenarios is higher for NO₂ and BC (reaching 40% in the urban background and 50% in the streets) than for other compounds, because road traffic is one of the main sources of those two pollutants in urban areas. The concentrations and their evolution under the different emission scenarios are not homogeneously distributed over Paris city. The left panel of Fig. 4 illustrates the average PM_{2.5} concentrations calculated in the 1-REF simulation, and the right panel, the NMB of PM_{2.5} concentrations between the 3-PET scenario and the 1-REF simulation. PM_{2.5} concentrations are the highest in the north of Paris, reaching 60 µg m⁻³ in streets with high traffic emissions, but only 23.6 µg m⁻³ in the urban background. The impact of the 3-PET scenario is also higher in regions with high traffic emissions. The decrease of the PM_{2.5} annual concentrations induced by the 3-PET scenario reaches 40% in the streets, while only 16% in the urban background.

As for PM_{2.5}, the concentrations of all analysed compounds are higher in the streets than in the urban background (see Figures 2, 3,

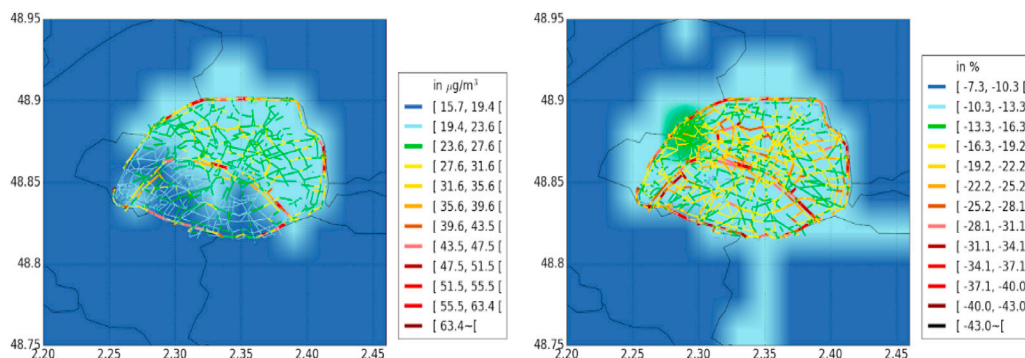


Fig. 4. Average annual concentrations of $PM_{2.5}$ calculated with the 1-REF simulation (left panel) and the NMB obtained by comparing the 3-PET scenario to the 1-REF simulation (right panel).

4, 5 and 6 in Supplementary Material section). The ratios between the maximum annual average concentration observed in the street-network and in the urban background ($f_{c_{st-bg}}$) are higher for PM_{10} (5.4), PM_{org} (6.2), $PM_{org,anth}$ (8.6) and BC (9.0) than for NO_2 (3.2), and $PM_{2.5}$ (2.7). This can be explained by the secondary formation of NO_2 and $PM_{2.5}$ above the streets from gas precursors emitted by traffic or other sources. Furthermore, even though road traffic is an important source of primary $PM_{2.5}$, it is also largely emitted by other sources, such as residential cooking and heating (Yun et al., 2020; Archer-Nicholls et al., 2016; Ou et al., 2020).

The impact of the scenarios is higher in the streets than in the urban background for all compounds. The ratios between the impact of the scenarios on concentrations in the streets and in the urban background ($f_{i_{st-bg}}$) are higher for $PM_{2.5}$ (2.6), PM_{org} (2.4) and PM_{10} (2.4), than for $PM_{org,anth}$ (1.8) and BC (1.2) and NO_2 (1.2). As road traffic is the main source of NO_2 , BC and $PM_{org,anth}$, the traffic reduction scenarios considerably decreased concentrations in both the streets and the urban background, resulting in lower values of $f_{i_{st-bg}}$. As $PM_{2.5}$, PM_{10} and PM_{org} have other main sources in the urban background, such as residential and biogenic emissions, the impact of the scenario is higher in the streets than in the urban background, resulting in higher values of $f_{i_{st-bg}}$. According to the AIRPARIF emissions inventory in 2015 (AIRPARIF, 2018), the residential sector is the main source of PM_{10} emissions in Île-de-France region (33%), more important than road traffic sector (26%). Other sectors, such as agriculture and construction sites, contribute with 17% and 16% of PM_{10} emissions, respectively.

The seasonal impact of the 3-PET scenario on urban air quality (NO_2 , $PM_{2.5}$, PM_{10} , PM_{org} , $PM_{org,anth}$, and BC) is quantified by the NMB, and shown in Figure 7, in Supplementary Material section. The purple and green columns indicate the NMB calculated in the streets and in the urban background, respectively. For each species, the first column corresponds to the annual average NMB, followed by the average NMB of winter, spring, summer and autumn. For all species and in all seasons the impact of the scenarios is higher in the streets than in the urban background. The NMB of BC is fairly constant at all seasons, because (i) it is mainly emitted by road traffic, which does not strongly vary during the year, and (ii) it is an inert compound, thus indifferent to changes in meteorological conditions. Higher differences between the seasons are observed for reactive compounds, such as NO_2 , $PM_{2.5}$ and PM_{org} . For NO_2 , the concentrations are slightly lower in summer (see Fig. 3 in the Methods section), but the impact of the scenarios is higher, suggesting that sources other than traffic are less important in summer. The opposite is observed for PM_{org} : summer is the season with the lowest impact of the scenarios. Biogenic emissions of organic compounds are higher in summer, as solar radiation is higher, increasing the organic concentrations of biogenic origin, and decreasing the relative importance of traffic emissions on total organic concentrations. For PM_{10} and $PM_{2.5}$, the impact of the scenarios is higher during

cold seasons (winter and autumn), as the cold temperatures favour the condensation of semi-volatile organic compounds emitted from road traffic.

Although the largest concentration differences are observed between the scenarios and the reference, there are non-negligible differences between the different scenarios. As observed by Andre et al. (2020) for background concentrations, the decreases of BC, $PM_{2.5}$, NO_2 and PM_{org} concentrations are larger by promoting recent petrol vehicles (3-PET scenario) than recent diesel vehicles (2-BAU scenario). Regarding the vehicle fleet evolution (2-BAU, 3-PET and 4-ELEC scenarios), the largest decrease of pollutant concentrations is observed by promoting electric vehicles: for NO_2 , the decrease is 50% in the 4-ELEC scenario, against 45% and 38% in the 3-PET and 2-BAU scenarios, respectively; and for $PM_{org,anth}$ the decrease is 34% in the 4-ELEC scenario, against 31% and 32% in the 3-PET and 2-BAU scenarios. The differences between the scenarios are lower for BC, $PM_{2.5}$ and PM_{10} because the differences of exhaust emissions are small, and non-exhaust emissions are the same between the fleet evolution scenarios (see the Methods section). For $PM_{2.5}$, all scenarios present a quite similar concentration decrease in the streets (ranging from 19% with the 2-BAU scenario, to 21% with the 4-ELEC scenario). The home-office practice (5-PET_{ho} scenario) does not lead to a drastic decrease of concentrations compared to the 3-PET scenario. However, it enhances the decrease of BC, PM_{10} and $PM_{org,anth}$ concentrations, as they are strongly impacted by non-exhaust emissions. The average decrease of BC is 54% with the 5-PET_{ho} scenario, against 51% with the original 3-PET scenario. The lowest concentrations are observed for the 5-PET_{ho} scenario for particles, and for the 4-ELEC scenario for NO_2 , even if the differences between scenarios are low. Note that the 4-ELEC scenario increases the electric vehicles proportion in the vehicle fleet, but it still adopts a relatively low percentage of electric light-duty vehicles (11%), as detailed in the Methods section. A higher fraction of electric vehicles should favour additional reductions NO_2 and $PM_{org,anth}$ concentrations. As mentioned in Section 2, even if these conclusions are based on simulations with a reference situation in 2014, they are still relevant in 2021/2022. In January 2021 the observed percentage of electric and hybrid vehicles in France is still low (less than 1%), non-exhaust emissions are similar and even higher than 2014 because the traffic flow has slightly increased (about 4.5%) and non-exhaust emissions are still not regulated. Furthermore, high concentrations of NO_2 , PM and BC are still often observed in Paris

5. Population exposure to multiple compounds

The impact of the scenarios on outdoor population exposure is assessed by crossing at the residential address, the population data (INSEE, 2019) and the pollutant concentrations, simulated either in the streets or in the urban background. Population in streets are defined by crossing the Parisian population density (km^2 resolution) with the

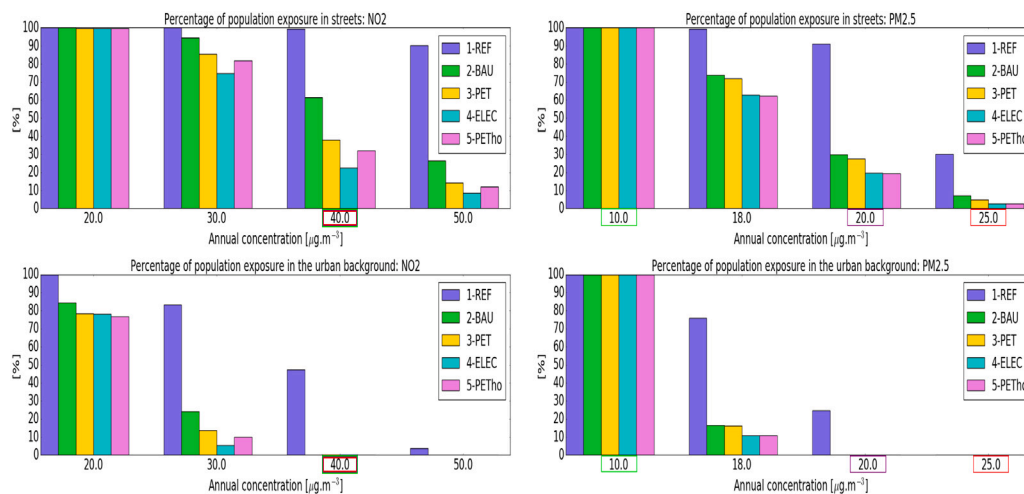


Fig. 5. Population exposure above threshold concentrations of NO_2 and $\text{PM}_{2.5}$ in the streets (top panel) and in the urban background (bottom panel).

coordinate of each street (starting and ending points). In case of more than one street observed in the same population grid, the population of each street is defined using a street length weighted average. Note that here only outdoor exposition is analysed. The “full” population exposure should also take into account indoor and commuting exposures (Languille et al., 2022). Fig. 2 illustrates the NO_2 and $\text{PM}_{2.5}$ outdoor population exposure using street or background concentrations (see the Extended data for other compounds). In the concentration axis, the green rectangle represents the air quality guidelines recommended by World Health Organisation for annual concentrations in force in 2014 (WHO, 2006), and the purple and red rectangles represent the limit value of annual concentrations from European Union and French regulations, respectively. The largest differences between the scenarios are observed for the population exposure to NO_2 . As expected, the population is exposed to higher concentrations near streets than in the urban background, stressing the importance of multi-scale simulations to evaluate the scenario efficiencies. All scenarios respect the French regulations for NO_2 and PM_{10} and the French guidelines for $\text{PM}_{2.5}$ if background concentrations are used to calculate population exposure, but it is not the case if exposure is calculated using street concentrations. While in the reference situation all Parisian population is exposed to higher concentrations than French/Europe/WHO guidelines ($40 \mu\text{g m}^{-3}$), this percentage decreases to 60% in the 2-BAU scenario, 38% in the 3-PET, 18% in the 4-ELEC, and 30% in the 5-PET_{ho}, benefiting at least 890 thousand inhabitants. For particles (BC, PM_{10} , $\text{PM}_{2.5}$, PM_{org} and $\text{PM}_{org,anth}$), the largest reductions of population exposure are observed for the 4-ELEC scenario, with similar reductions as the 5-PET_{ho} scenario. In the 1-REF simulation 90% of the population are exposed to $\text{PM}_{2.5}$ concentrations higher than the French guideline values ($20 \mu\text{g m}^{-3}$), representing about 1.99 million inhabitants. This percentage decreases to around 30% in the 2-BAU and 3-PET scenarios and reaches 20% in the 4-ELEC and 5-PET_{ho} scenarios, benefiting about 1.33 and 1.55 million inhabitants, respectively (see Fig. 5).

The decrease of population exposure is not homogeneous over the city. Figure 9 illustrates the NMB obtained by comparing the $\text{PM}_{2.5}$ concentrations simulated with the 3-PET scenario to those simulated with the 1-REF simulation in each street. Similar spatial distributions are obtained for PM_{10} , NO_2 , PM_{org} , $\text{PM}_{org,anth}$ and BC, and also for the other scenarios. Actions aiming at reducing traffic emissions should be prioritized in the regions where the NMB is the highest. Note that, according to simulation results in the streets, 100% of Parisian population is exposed outdoor to annual-average concentrations of NO_2 , $\text{PM}_{2.5}$ and PM_{10} higher than those recommended in 2005 by WHO. WHO guidelines were recently updated (World Health Organization et al., 2021), and the recommended annual concentration of NO_2 was

reduced from $40 \mu\text{g m}^{-3}$ to $10 \mu\text{g m}^{-3}$, and from $10 \mu\text{g m}^{-3}$ to $5 \mu\text{g m}^{-3}$ for $\text{PM}_{2.5}$, and from $20 \mu\text{g m}^{-3}$ to $15 \mu\text{g m}^{-3}$ for PM_{10} . This makes even more clear the need to reduce pollutant concentrations in urban areas, and the need of more ambitious reductions in pollutant emissions.

6. Conclusions

This study investigates the influence of vehicle fleet composition and urban mobility on the residential exposure of the Parisian population to outdoor NO_2 , $\text{PM}_{2.5}$, PM_{10} , BC and $\text{PM}_{org,anth}$ using an innovative approach, allowing to take into account the concentrations of multi-pollutants, including secondary aerosol formation. A systematic multi-pollutant assessment of outdoor exposure is complex, and it is often estimated for a limited number of pollutants, or using models with strong modelling assumptions (e.g. only background concentrations are considered, or secondary pollutants are ignored at the local scale). Here a multi-scale approach is used, one-way coupling the 3D Eulerian CTM Polair3D (regional scale) and MUNICH (local scale) to calculate pollutant concentrations in the urban background and in streets. Both models integrate the chemical module SSH-aerosol, and this multi-model chain is to our knowledge the only chain capable of representing particle dynamics including the formation of organic and inorganic secondary particles in the urban background and in streets. Residential exposure to outdoor air pollution is estimated using a static approach, by crossing the residential address, the population data and the pollutant concentrations, simulated either in the streets or the urban background. Residential exposure to outdoor air pollution is higher in streets than in the urban background, not only because of the higher concentrations but also because of the particle chemical composition. As shown by 10-year evolution scenarios, renewal of the fleet technological composition and road mobility are clearly efficient to reduce air pollutant concentrations. All emission scenarios showed a strong reduction of population exposure to regulated (NO_2 , PM_{10} and $\text{PM}_{2.5}$) and non-regulated pollutants (BC, PM_{org} and $\text{PM}_{org,anth}$). The main air-quality improvements are obtained by promoting electric vehicles. The home-office practice enhances the decrease in particle concentrations induced by vehicle fleet renewal, but its impacts are low if used as a single measure. Still, none of the scenarios studied in this framework was able to avoid outdoor exposure to concentrations above the European/French and WHO air-quality limit/guideline values. Furthermore, as the updated WHO guidelines [WHO, 2021] were recently updated with even more strict air-quality recommendations, the need to reduce pollutant concentrations in Paris is evident. The 10-year evolution scenarios studied here were based on the year 2014. Even if non-negligible differences exist regarding the vehicle technologies

and Euro standards in the traffic fleet observed in Paris in 2014 and nowadays, these scenarios remain relevant, as the total vehicle fleet flow is similar. Furthermore, with the low fraction of electric vehicles in the current fleet (less than 1% compared to 11% in the scenario 4-ELEC), the scenarios presented here still seem ambitious. The strategy of vehicle fleet renewal seems insufficient to protect the whole Parisian population, and suggest the need for more ambitious measures. Further reductions of anthropogenic emissions can be proposed, such as controlling heating and cooking emissions from the residential sector. However, actions to reduce traffic-related emissions are needed, such as those currently advocated: increasing the proportion of recent and electric vehicles, setting low-emission zones, but also controlling non-exhaust emissions and changing urban mobility. International commitments, such as those of the 26th UN Climate Change Conference of the Parties (COP26) Declaration on Accelerating the Transition to 100% Zero Emission Cars and Vans (United Nations, 2021), are very important for climate issues and to reduce the outdoor population exposure to pollutants emitted by traffic exhaust, especially nitrogen oxides. However, even for a completely electrified fleet, the impact of traffic on outdoor population exposure in cities may still be high, because of non-exhaust traffic emissions. Recent studies indicates that electric vehicles may even emit more non-exhaust emissions than classic diesel and petrol powered vehicles, as they are often heavier (Timmers and Achten, 2016). Therefore, in terms of outdoor population exposure and air quality, the vehicle weight and non-exhaust emissions should also be considered when defining regulations and mobility.

CRedit authorship contribution statement

Lya Lugon: Conceptualization, Software, Visualization, Validation, Formal analysis, Writing – original draft. **Youngseob Kim:** Software. **Jérémy Vignerot:** Resources. **Olivier Chrétien:** Funding acquisition. **Michel André:** Conceptualization. **Jean-Marc André:** Conceptualization, Resources. **Sophie Moukhtar:** Conceptualization, Resources. **Matteo Redaelli:** Conceptualization, Resources. **Karine Sartelet:** Conceptualization, Software, Formal analysis, Resources, Funding acquisition, Writing – original draft.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Statistical indicators

A.1. Indicators for model evaluation

- Fractional bias: $FB = 2 \left(\frac{\bar{o}-\bar{c}}{\bar{o}+\bar{c}} \right)$
- Geometric mean bias: $MG = \exp(\ln(\bar{o}) - \ln(\bar{c}))$
- Normalized mean square error: $NMSE = \frac{(\bar{o}-\bar{c})^2}{\bar{o}\bar{c}}$
- Geometric variance: $VG = \exp[(\ln(\bar{o}) - \ln(\bar{c}))^2]$
- Normalized absolute difference: $NAD = \frac{|\bar{o}-\bar{c}|}{(\bar{c}+\bar{o})}$

- Fraction of predictions within a factor two of observations: $0.5 \leq \frac{c}{o} \leq 2.0$

where o and c represent the observed and simulated concentrations respectively.

A.2. Indicators for model sensitivities

Normalized mean bias and error:

$$NMB = \frac{\overline{(c_1 - c_0)}}{\bar{c}_0}; \quad NME = \frac{\overline{|c_1 - c_0|}}{\bar{c}_0}$$

where c_0 and c_1 are the simulated concentrations in the reference and in the sensitivity simulations respectively.

Appendix B. Supplementary data

More information about simulation validation and impact of emission scenarios on concentrations and population exposure.

Supplementary material related to this article can be found online at <https://doi.org/10.1016/j.apr.2022.101365>.

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