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Mobile measurements and street-level modelling to assess outdoor and indoor personal exposure to air pollution in urban environment

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Abstract

Population exposure to atmospheric pollution is commonly assessed through models. This study aims to compare estimates based on a high-resolution model and actual measurements. The considered modelling system, ATMO-Street, consists of a three-layer outdoor model operating at an hourly rate with a spatial resolution of approximately 10 m. The considered measurements were made by 38 candidate citizens carrying, for 1 week, portable devices, including an AE51 aethalometer and an Antilope low-cost sensor system developed at the Scientific Institute of Public Service (ISSeP). Their data were aggregated to match the model time and space resolutions. The zone of interest is the city of Liège in Belgium during parts of the year 2019. This research sheds light on the effectiveness of the atmospheric pollution model and personal exposure assessment methods. The findings contribute to a somewhat more comprehensive understanding of our exposure to air pollution, including indoors, with potential implications for public health and environmental policy.

Keywords Personal exposure · Mobile measurement · Low-cost sensor · Street-level modelling · Particulate matter · Black carbon

Introduction

Atmospheric pollution with its adverse effects on health (World Health Organisation 2023) remains an issue for most of the European cities (EEA 2023). The limited number of monitoring stations located in urban environments usually prevents one to understand the various sources of pollution and to capture its full spatial variability. Air quality models are thus valuable tools to evaluate the spatiotemporal variability of pollution at high resolution. The combination of simulated pollution and population density maps provides the population exposure maps, which reflect how many people breathe a certain quantity of atmospheric pollutants. Some major simplifications are still often made in many exposure studies due to a lack of available scientific data. First, exposure estimates typically assume static population using population density maps, which are generated on the basis of residency information taking the population activities not into account. Secondly, they do not consider the indoor air contribution despite the fact that we spend most of our time indoors. A study of the United States Environmental Protection Agency (EPA) shows that, on average, Americans spend 87% of their time in a building, 6% in a vehicle and 7% outdoors (Klepeis et al. 2001). Similarly, Dons et al. (2012) showed that, on average, Belgian participants in their study spent 81% of their time in a building and 6% in a vehicle. Unfortunately, the researches focusing on time-activity pattern remain rare while the estimation of the time spent by individuals in various microenvironments is a critical aspect of exposure assessment to air pollution.

We now have the tools to overcome these limitations and give a much more accurate estimate of exposure, thanks in particular to the advent of portable sensors and the rise of citizen science (EEA 2019). Numerous studies have already been carried out using portable sensors (Alas et al. 2019; Deshmukh et al. 2020; Elen et al. 2013; Kaur et al. 2006; Van Poppel et al. 2023). Most of these studies were dedicated to the monitoring of outdoor air pollution on the roads using cars (Apte et al. (2017) for nitrogen oxides (NO_x) and black carbon (BC)) or bicycles (Int Panis et al. (2010) for particulate matter (PM_x), Peters et al. (2014) for ultrafine

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particles (UFP) and BC, Van den Bossche et al. (2015) for BC). Studies also recently focused on the deep validation of air quality models, like the state-of-the-art ATMO-Street model, using trajectory measurements or citizen-dense networks (Vandeninden et al. 2021; Hooyberghs et al. 2022).

In this context, the objective of this study is double. The first part consists in assessing the personal outdoor and indoor exposure to $PM_{2.5}$ (the fraction of particles smaller than 2.5 µm) and BC pollution. This is done by providing candidate citizens of Liège with a homemade portable low-cost sensor system (LCSS) and an AE51 aethalometer. The second part considers the use of the mobile measurements recorded during travels and outdoor activities to evaluate the ATMO-Street model capacity to simulate the high spatiotemporal variability of concentrations or at least to compare its output against mobile measurements.

The paper is divided into three sections. In the "Materials and methods" section, we present the portable sensors we used for mobile measurements, notably the ISSeP LCSS, as well as the ATMO-street air quality model chain and its set-up for the Liège area. In the "Results" section, we summarize the exposure measurements to $PM_{2.5}$ and BC and the comparison with model results. In the "Discussion" section, we comment the uncertainties and limitations linked to the different approaches.

Materials and methods

Measurements

In the framework of the Outdoor and Indoor Exposure (OIE) project, mobile measurement campaigns carried out by citizens were carried out in 2019 in the city of Liège to explore personal exposure to atmospheric pollutants. Liège is located in the eastern part of Belgium and lies, in the Meuse Valley, close to the borders with the Netherlands and Germany. It has a population of around 200,000 people.

We provided each participant with a set of portable devices: a portable low-cost sensor system (LCSS) for air quality monitoring (Lenartz et al. 2021), designed by ISSeP with its partner HEPL (Haute École de la Province de Liège), a portable AethLabs AE51 aethalometer for the measurement of black carbon (BC) and a GlobalSat DG200 GPS to track the subject position. The ISSeP's LCSS, named Antilope, is made up of a Sensirion SPS30 optical sensor for the measurement of $PM_{2.5}$, Alphasense electrochemical sensors for nitrogen oxides (nitrogen oxide NO and nitrogen dioxide NO_2) and ozone (O_3) approximate levels and a BME680 sensor for the measurement of temperature, humidity and pressure. See Lenartz et al. (2021) for a full description of the equipment and an evaluation of its performance. In the present paper, we focus only on particulate matter and, more specifically, on the fraction of particles smaller than 2.5 µm ($PM_{2.5}$) and black carbon (BC), because they require none or simple correction to be in good agreement with reference analyzers, contrary to gaseous pollutants. As already shown by Hofman et al. (2018), uncertainties related to AE51 measurements are relatively small. All the BC measurement data were processed using the optimised noise-reduction algorithm (Hagler et al. 2011).

Measurements have been almost continuously performed for both $PM_{2.5}$ and BC from April 24 to August 7 and from November 12 to December 9 in 2019 by 38 participants in Liège (four per week). A candidate citizen relayed one other receiving all the equipment for a week, in a backpack to easily shadow her/him in her/his daily activities. ISSeP sensor system was set at a 1-s record rate and the measurements were then aggregated by minute to correspond to the temporal resolution of black carbon and location data. In total, there are more than 3000 h of data collected. Every day during the week, participants had to fill in a daily logbook with all their activities. Each activity had to be characterized by a start time, an end time, a type (work, shopping, staying at home, cooking, sport, leisure, etc.) and an environment (indoor or outdoor). Travels are considered an activity with an indoor/outdoor type according to the mode of transport (car, bus, train, walk, etc.).

Modelling

High-resolution $PM_{2.5}$ and BC concentrations were simulated over the city of Liège using the ATMO-Street quality model chain (Lefebvre et al. 2013a). ATMO-Street combines the Immission Frequency Distribution Model (IFDM, Lefebvre et al. 2013b), a bi-Gaussian plume dispersion model for the dispersal of local industrial and traffic emissions and the Operational Street Pollution Model (OSPM, Berkowicz et al. 2008), simulating the specific dispersion of pollutants within street canyons. In the city centre, most of the streets, classified as street canyons, are simulated by the OSPM model. Pollutant concentrations are calculated at various points (receptors) in the study area and then interpolated for the whole zone at a 10-m spatial resolution. Model simulation was performed over the year 2019 at an hourly rate.

To determine the urban background concentrations, we used the measurements of the reference monitoring station located in Herstal (station TMLG05, 50.658416° N, 5.627990° E; Fig. 2), a suburban area of Liège (https://www.wallonair.be). The 2019 annual mean background concentrations in BC and $PM_{2.5}$ are respectively 0.8 $\mu g m^{-3}$ and 8.6 $\mu g m^{-3}$. We considered the most important industries in the area as local emission point sources. There are 31 of them with hourly emissions from 10 g to 3.85 kg for $PM_{2.5}$ and from 10 to 110 g for BC (see their location in Figure S1 of the Supplementary Material). The BC and $PM_{2.5}$ line

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Journal : Large 11869 | Article No : 1529 | Pages : 12 | MS Code : 1529 | Dispatch : 31-1-2024

emissions corresponding to the traffic were estimated using the HERE floating car data for 2018 (Figures S1 and S2 of the Supplementary Material). The traffic flows were converted into emissions using the vehicle fleet of the Walloon Region in 2019 and the emission factors from COPERT database (https://www.emisia.com). The total BC and $PM_{2.5}$ emissions from traffic for an average day in Liège are respectively 144 kg and 368 kg (traffic daily variation in Table S1 of the Supplementary Material). The emissions of other sectors (residential, agriculture, etc.) are not taken into account by the model.

We used local stations to set the meteorological conditions in 2019: the ISSeP Saint-Nicolas station (TMSG02, Fig. 2) for temperature and wind (speed and direction) and the Pameseb Alleur station for solar radiation (50.682404° N, 5.494508° E). A wind rose for 2019 is presented in Figure S3 of the Supplementary Material.

Comparison of measurements and modelling output

In order to validate a modelling system, one needs to compare its output against data with a similar representativeness. In the present case, ATMO-Street has a temporal resolution of 1 h and a spatial resolution of about 10 m, whereas the measurements are 1-min averaged values and represent the average concentration at one specific location or along a track corresponding to a 1-min displacement. The covered distance in this time interval could be up to 90 m for a pedestrian (walking at 6 km/h) and about 833 m for a car (driving at 50 km/h); in both cases, the trajectory is mapped over more than one model grid cell. Furthermore, the modelling system provides only outdoor concentrations, which cannot be directly related to the measurements recorded inside a vehicle. In addition, it is known that the concentrations can vary largely depending on whether the measurements are made in the middle of the road, on the kerbside or close to the building façades. OSPM, the street-canyon component of ATMO-Street, computes the pollutant concentrations right in front of the buildings, while pedestrians, cyclists and bikers can move across the whole width of the sidewalk and the street.

Hence, for each hour and each grid cell for which data are available, we compute the mean of all outdoor measurements that are geolocated within the same considered grid cell and timestamped within the same considered period. This approach still has its limitations (e.g. regarding the subject exact location) but makes the comparison of measurements and modelling output more fair. Because of these discrepancies in terms of representativeness, we should rather talk about comparison than thorough validation.

Results

Thirty-eight citizens made both 1-min rate BC and $PM_{2.5}$ measurements during their indoor and outdoor daily activities in 2019 in Liège. The activity frequency and associated exposures are presented in Table 1. The participants spent more than 80% of their daytime indoors (48% at home and 23% at work, Table 1) and in daily commutes (12%). In comparison, the time spent outside when people are at home or at work corresponds only to 5% of the daily activities. The remaining time is essentially shared between leisure (5%),

AQ7 **Table 1** Indoor and outdoor daily activities with their frequency and exposure (median (p_{50}) and the 90th percentile (p_{90})) to particle matter ($PM_{2.5}$) and black carbon (BC) (average of 38 participants)

Activity	Duration (h)	Frequency (%)	$PM_{2.5}$, p50 (μgm^{-3})	$PM_{2.5}$, p90 (μgm^{-3})	BC, p50 (μgm^{-3})	BC, p90 (μgm^{-3})
Indoor home	1648	48.2	6	35	0.6	1.7
Indoor work	797	23.3	4	10	0.4	1.2
Travels	424	12.4	4	17	0.9	3.7
Outdoor home	113	3.3	8	18	0.7	2.4
Indoor leisure	88	2.6	6	75	0.5	3.6
Outdoor leisure	80	2.4	5	16	0.8	3.1
Cooking	77	2.3	7	51	0.7	2.7
Shopping in the city	68	2.0	5	16	0.9	3.7
Outdoor work	52	1.5	6	11	0.8	3.3
Shopping in a mall	25	0.7	4	16	0.7	2.3
Outdoor sport	16	0.5	14	28	0.8	3.0
Indoor sport	15	0.4	9	90	0.5	1.7
Picking up children from school	11	0.3	6	12	0.9	4.0
Making a fire	5	0.1	4	15	1.3	6.1

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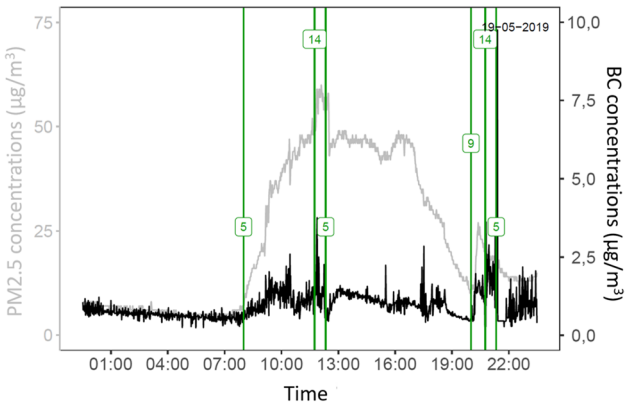
Journal : Large 11869 | Article No : 1529 | Pages : 12 | MS Code : 1529 | Dispatch : 31-1-2024

shopping (3%), cooking (2%) and sport (1%). The proportion of these activities performed indoor or outdoor depends of course on the season (e.g. shopping in the city or in a mall). An example of daily activity pattern and associated $PM_{2.5}$ and BC exposure is presented in Fig. 1.

To outline the results of this personal exposure campaign, it seemed to us more pertinent to present the median (or 50th percentile) and the 90th percentile (p_{90}) of measured concentrations, rather than the mean and the maximum, because people can sometimes be very close to pollution source during their activities and on their routes (Table 1). The median exposure to $PM_{2.5}$ is equal to 6 μgm^{-3} at home and to 4 μgm^{-3} at work and during commutes. It is higher when people are cooking (7 μgm^{-3}), are outdoor at home (8 μgm^{-3}) or are doing sport (9 μgm^{-3} indoors and 14 μgm^{-3} outdoors). It is during these indoor activities that the highest concentrations were recorded. The value of the 90th percentile is 90 μgm^{-3} for indoor sport, 75 μgm^{-3} for other indoor leisure, 51 μgm^{-3} for cooking and 35 μgm^{-3} when being simply at home. Concerning the exposure to BC, the campaign attests this pollutant is a good indicator of traffic and more generally of combustion. Considering the median concentrations, people are the most exposed to BC when making a fire (1.3 μgm^{-3} and p_{90} value = 6.1 μgm^{-3}), ahead of leaving and picking up children from school (0.9 μgm^{-3} and p_{90} value = 4.0 μgm^{-3}), travelling (0.9 μgm^{-3} and p_{90} value = 3.7 μgm^{-3}) or shopping in the city (0.9 μgm^{-3} and p_{90} value = 3.7 μgm^{-3}).

To have an idea of the mean daily exposure, we weighted the exposure of our population sample during the different types of activities by the time spent for each. The participants were in average exposed to 5 μgm^{-3} in $PM_{2.5}$ and 0.6 μgm^{-3} in BC during their daytime (17 μgm^{-3} in $PM_{2.5}$ and 1 μgm^{-3} in BC if the mean values are used). If we only consider the indoor activities, the median exposure remains the



AQ8 **Fig. 1** Daily pattern of activities and associated $PM_{2.5}$ and BC exposure of a participant in Liège. Green lines represent the start of an activity (5 = indoor home, 14 = travel and 9 = cooking)

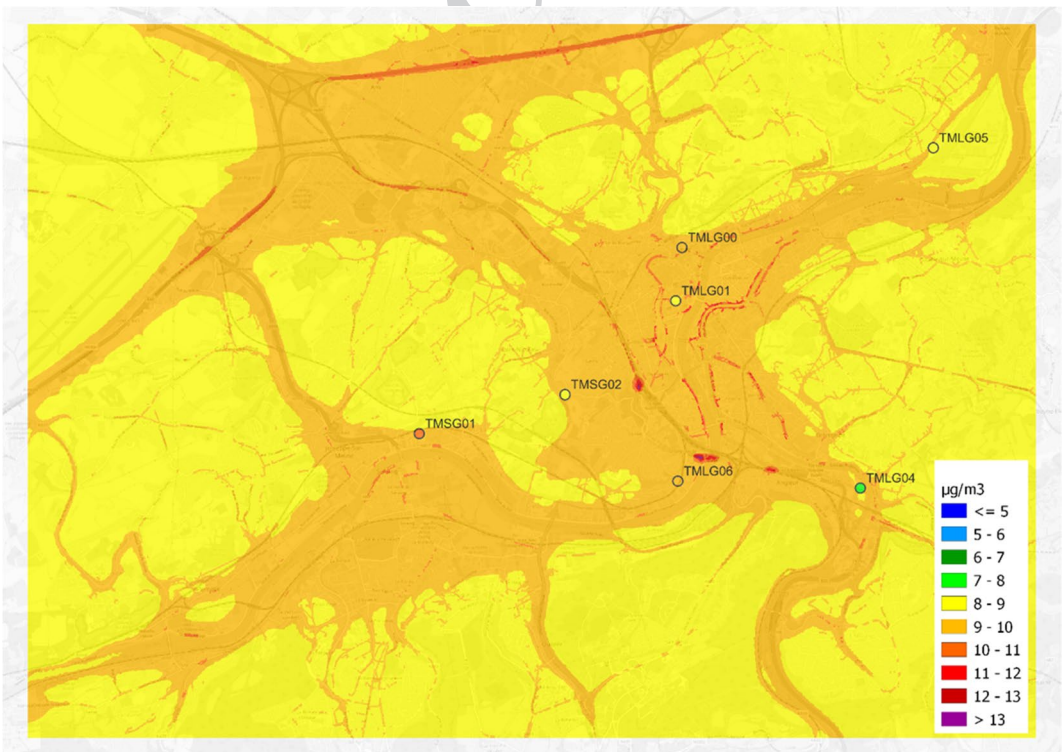


Fig. 2 $PM_{2.5}$ particle matter annual mean concentrations simulated by the ATMO-Street model (10-m spatial resolution) and measured at regional stations (circles) in Liège in 2019

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Journal : Large 11869 | Article No : 1529 | Pages : 12 | MS Code : 1529 | Dispatch : 31-1-2024

same but the mean exposure to $PM_{2.5}$ and BC, respectively, increase (18 μgm^{-3}) and decrease (0.9 μgm^{-3}) a little bit. During their daily outdoor activities, they were exposed to 5 μgm^{-3} in $PM_{2.5}$ and 0.8 μgm^{-3} in BC (11 μgm^{-3} in $PM_{2.5}$ and 1.5 μgm^{-3} in BC if the mean values are used).

Before analysing outdoor personal exposure in the light of the ATMO-Street model, we compared the model outputs with the reference stations of the regional air monitoring network located in the Liège municipality (seven $PM_{2.5}$ and two BC monitoring locations). Figure 2 displays the simulated and measured $PM_{2.5}$ annual mean concentrations for 2019 (Figure S4 of the Supplementary Material for BC concentrations). If we consider the ISSeP traffic station (TMLG01), located in the city centre (50.637978° N, 5.571781° E, Fig. 2), the 2019 annual mean concentrations in BC and $PM_{2.5}$ are respectively 1.05 μgm^{-3} and 8.5 μgm^{-3} for the reference station and 1.08 μgm^{-3} and 9.4 μgm^{-3} according to the model (see Figures S5 et S6 for the daily concentrations over 2019). Comparisons with the other reference stations are available in Table S2 of the Supplementary Material. The same comparison with mobile measurements is impossible owing to few measurements available in space and in time close to the station. We also estimated the exposure of Liège population in 2019 using the regional reference stations. We attributed to each 10 m × 10 m grid cell of the territory the annual mean (not the median) concentration value of the closest station. The annual mean exposure of the Liège population was then determined by weighting the concentrations by the 100-m population. It results in an estimation of population exposure (8 μgm^{-3} for $PM_{2.5}$ and 1 μgm^{-3} for BC) lower than personal exposure derived from mobile measurements (11 μgm^{-3} for $PM_{2.5}$ and 1.5 μgm^{-3} for BC). Using similarly the 10-m ATMO-Street concentrations, we got a mean exposure of 9 μgm^{-3} for $PM_{2.5}$ and 1 μgm^{-3} for BC in 2019.

Outdoor exposure, during daily travels in particular, was analysed with more attention using mobile measurements and street-level air quality model in parallel. To realize this, the 1-min pollution measurements were aggregated over the period of the campaign (April 24 to December 9) at the 10-m spatial resolution of the model. $PM_{2.5}$ and BC measured and modelled concentrations for travels and outdoor activities are displayed in Table 2. Regarding the modal split, half of the daily trips are made by car (53%), followed by walking (24%), train (8.5%), bus and bike (both equal to 7%). Similarly to activities, the travel mode choice depends on the meteorological conditions. Even if $PM_{2.5}$ concentrations are not really the best marker to study exposure to traffic, some elements stand out. According to measurement and model, pedestrians (p_{90} value = 25 μgm^{-3}) and bus commuters (p_{90} value = 21 μgm^{-3}) are the most exposed to $PM_{2.5}$ pollution. The median concentration for cyclist is identical, but p_{90} value is lower (12 μgm^{-3}). Car drivers are less

Table 2 Comparison between $PM_{2.5}$ and BC measured (MEAS) and modelled (ATMO) concentrations for travels and outdoor activities (ranked by daily frequency). Statistics used to compare data and model values are the median (p_{50}), the 90th percentile (p_{90}), the Pearson correlation coefficient (r) and the root-mean-square error (RMSE)

Activity	Daily frequency (%)	$PM_{2.5}$ (μgm^{-3})							BC (μgm^{-3})						
		MEAS, p50	ATMO, p50	MEAS, p90	ATMO p90	r	RMSE		MEAS, p50	ATMO, p50	MEAS, p90	ATMO, p90	r	RMSE	
Travels	12.4	4	5	17	20	0.23	26.9		0.9	0.9	3.7	2.1	0.11	4.1	
Car	52.6	3	5	13	25	0.25	13.4		1.1	1.0	4.4	2.0	0.14	3.4	
Walk	23.7	6	6	25	25	0.27	46.0		0.6	0.8	3.0	2.7	0.06	5.9	
Train	8.5	3	4	8	7	0.24	9.7		0.8	0.9	3.0	1.8	0.32	1.7	
Bus	6.7	6	8	21	20	0.33	14.5		1.4	1.0	4.2	2.2	0.25	2.5	
Bike	6.6	6	5	12	14	0.83	3.5		0.7	0.9	2.0	1.2	0.10	3.3	
Moto	1.9	5	2	10	15	0.91	4.1		0.7	0.7	1.6	1.5	0.36	0.9	
Outdoor home	3.3	8	8	18	30	0.53	8.7		0.7	1.0	2.4	2.1	0.28	1.0	
Outdoor leisure	2.4	5	3	16	8	0.77	5.6		0.8	0.9	3.1	1.3	0.11	1.5	
Shopping in the city	2.0	5	6	16	24	−0.03	17.4		0.9	1.0	3.7	2.5	0.05	10.8	
Outdoor work	1.5	6	3	11	14	0.59	3.9		0.8	0.6	3.3	1.6	0.11	3.8	
Outdoor sport	0.5	14	8	28	19	0.15	12.2		0.8	0.9	3.0	1.2	-0.21	1.2	
Picking up children from school	0.3	6	10	12	19	−0.11	29.9		0.9	1.2	4.0	1.7	0.29	2.6	

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Journal : Large 11869 | Article No : 1529 | Pages : 12 | MS Code : 1529 | Dispatch : 31-1-2024

exposed according to the data (3 μgm^{-3}) while, with the model, car drivers are as exposed as pedestrians are. Measurement and model also agree on the low exposure when taking the train (lowest p_{50} and p_{90} values). There are too few data to conclude something for motorbikes. If median mobile data and model values are equal for walkers, it is for this mode that the root-mean-square error (RMSE) is the highest (46 μgm^{-3}).

For outdoor activities, the model tends to simulate lower median concentrations (3 μgm^{-3}) compared to what the data indicate, for the activities supposedly further from the road (e.g. leisure or work). For activities closer to the roads, the model results are higher (e.g. 6 μgm^{-3} for shopping in the city and 10 μgm^{-3} for picking up children from school). Data and model agree on quite high median concentrations of 8 μgm^{-3} for outdoor home activities. Concentrations are also particularly high during outdoor sport (14 μgm^{-3} for data and 8 μgm^{-3} for model) but it only corresponds to a few measurement hours. Pearson correlation coefficients (r) between measurement and model are not strong, except for travels by bike and outdoor home activities but only few measurements characterized these both categories. Correlation is moderate for some other outdoor activities (but slightly negative for shopping in the city and picking up children with high RMSE) and weak for other trip modes.

As regards exposure to BC, both methods point out that bus commuters and car drivers are exposed to the highest concentrations (respectively 1.4 μgm^{-3} and 1.1 μgm^{-3} for data and 1 μgm^{-3} for model). The less exposed are the walkers (0.6 μgm^{-3} for data and 0.8 μgm^{-3} for model), ahead of bike (0.7 μgm^{-3} for data and 0.9 μgm^{-3} for model) and train (0.8 μgm^{-3} for data and 0.9 μgm^{-3} for model); differences between modes are being less pronounced with the model. The correlations between measurement and model are all weak and, similarly to $PM_{2.5}$, the RMSE is high for concentrations during walk trips. Concerning outdoor activities, the model simulates higher median concentrations than measured by the citizens (except for outdoor work) but the dataset always displays higher p_{90} values.

Figure 3 illustrates Table 2 but also highlights the high variability in $PM_{2.5}$ and BC concentrations measured by citizens during their daily commutes compared to the concentrations simulated by the model. Although the mobile measurements were aggregated to match the lower spatiotemporal resolution of the model outputs, they remain impacted by their initial 1-min resolution that allows to capture particular events or sources (e.g. traffic congestion, vehicle start-up or street food) that are not considered by the model. For the graph visibility, the $PM_{2.5}$ and BC concentration scales have been limited respectively to 100 μgm^{-3} and 10 μgm^{-3}. For $PM_{2.5}$, the maximum recorded concentrations are 193 μgm^{-3} for car, 802 μgm^{-3} for walk, 134 μgm^{-3} for train, 168 μgm^{-3} for bus, 28 μgm^{-3} for bike and 12 μgm^{-3} for motorbike. For BC, the maximum recorded concentrations are 47 μgm^{-3} for car, 190 μgm^{-3} for walk, 13 μgm^{-3} for train, 23 μgm^{-3} for bus, 51 μgm^{-3} for bike and 7 μgm^{-3} for motorbike.

Figures 4 and 5 show, respectively for $PM_{2.5}$ and BC, the spatial distribution of the concentration measurements corresponding to outdoor activities and travels, aggregated over the all measurement periods at the model 10-m resolution. In both Figs. 4a and 5a, we can see a walking tour during which one of the participants was clearly exposed to high concentrations in $PM_{2.5}$ and BC. For $PM_{2.5}$ concentrations, as displayed in Table 2, the model seems to overestimate the car driver exposure even if, in some points, like highways, modelled concentrations are lower than measured ones (Fig. 4b). For pedestrians, the model tends to overestimate $PM_{2.5}$ pollution in streets with heavy traffic and to underestimate it in quieter streets or pedestrian zones (Fig. 4c). More correlated with traffic, peaks in BC concentrations are observed in the measurement data on the highways and national roads (Fig. 5a) contrary to $PM_{2.5}$ map (Fig. 4a). There is no apparent pattern in the comparison between modelled and measured BC concentrations for cars. Along the same road, the model sometimes overestimates and sometimes underestimates concentrations. In contrast, the BC exposure of walkers is overestimated almost everywhere by the model (Fig. 5c). A well-known issue is that the model tends to overestimate concentrations along the river borders and large boulevards. We can also see that pedestrians and car drivers do not use the same routes. That can explain why walkers or cyclists, using low-traffic or car-free lanes, are less exposed to BC than conductors according to both measurement and model.

Discussion

Participants spent nearly 80% of their daytime indoors. They passed half of their time at home where they are exposed to median $PM_{2.5}$ and BC concentrations of 6 μgm^{-3} in $PM_{2.5}$ and 0.6 μgm^{-3}, corresponding to mean daily exposure calculated by weighting all the different activity exposure by elapsed time. For $PM_{2.5}$, the mean daily exposure is higher than the new World Health Organization (WHO) guideline of 5 μgm^{-3}. Workers are less exposed during working hours than at home (4 μgm^{-3} in $PM_{2.5}$ and 0.4 μgm^{-3}). It is during indoor activities that the highest $PM_{2.5}$ concentrations (i.e. 90th percentile of measured concentrations) are recorded (75 μgm^{-3} during indoor leisure, 51 μgm^{-3} when cooking or 35 μgm^{-3} when being simply at home).

During their daily outdoor activities, they were exposed to 5 μgm^{-3} in $PM_{2.5}$ and 0.7 μgm^{-3} in BC. Concerning personal exposure during daily commutes, our dataset suggests that car drivers are less exposed to $PM_{2.5}$ than the other travel modes. For exposure to BC, the bus commuters are the

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Journal : Large 11869 | Article No : 1529 | Pages : 12 | MS Code : 1529 | Dispatch : 31-1-2024

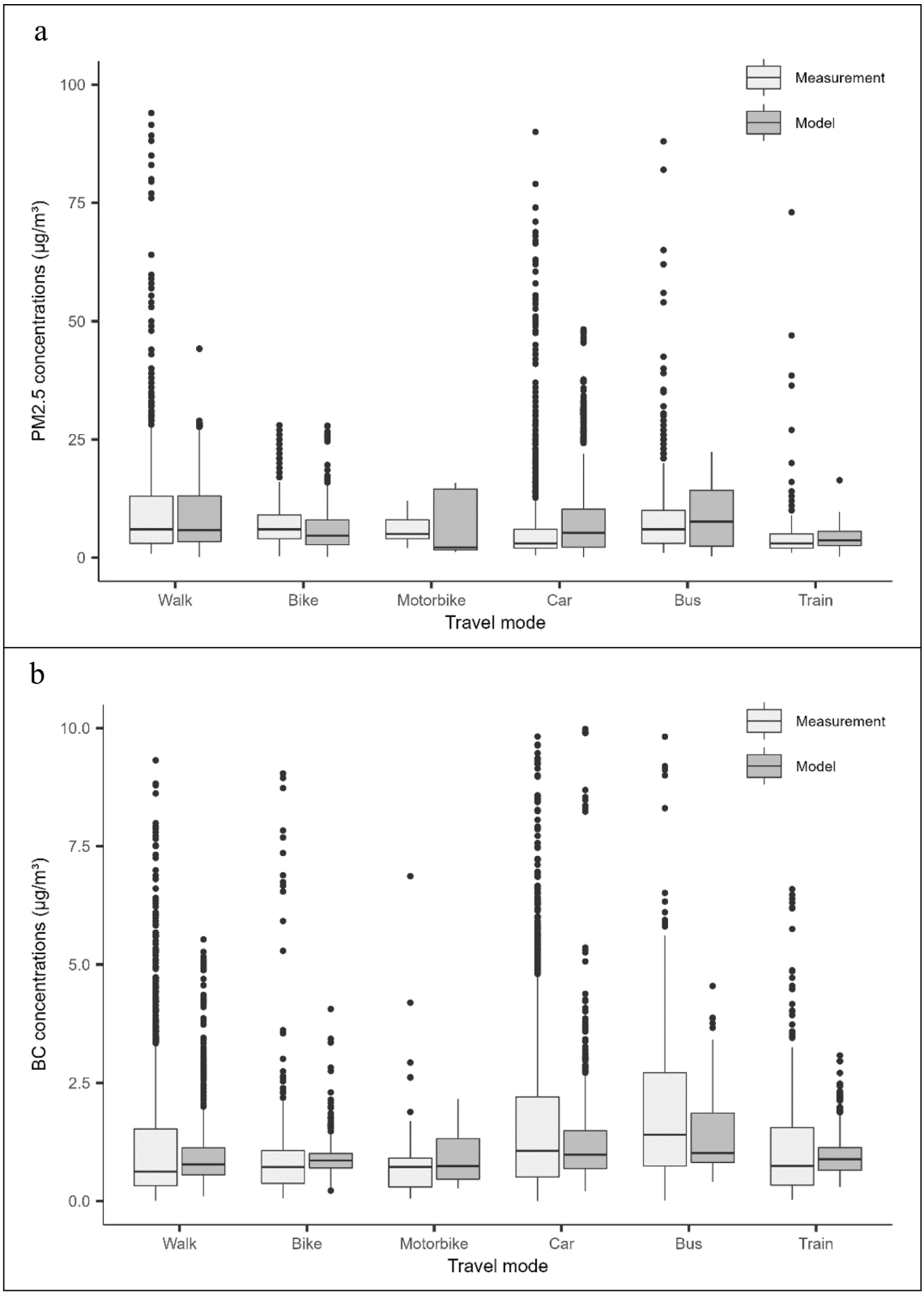

Fig. 3 Box plots summarising the distribution of $PM_{2.5}$ and BC concentrations measured by citizens or modelled by ATMO-Street for the different travel modes. The median is represented by the line in the box. The interquartile range (50% of the data) is represented by the box. The whiskers extending from both sides of the box represent the lower and upper 25% ranges of the values. For the graph visibility, the $PM_{2.5}$ and BC concentration scales have been limited respectively to 100 μgm^{-3} and 10 $\mu gm^{-}.^{3}$

Journal : **Large 11869** | Article No : **1529** | Pages : **12** | MS Code : **1529** | Dispatch : **31-1-2024**

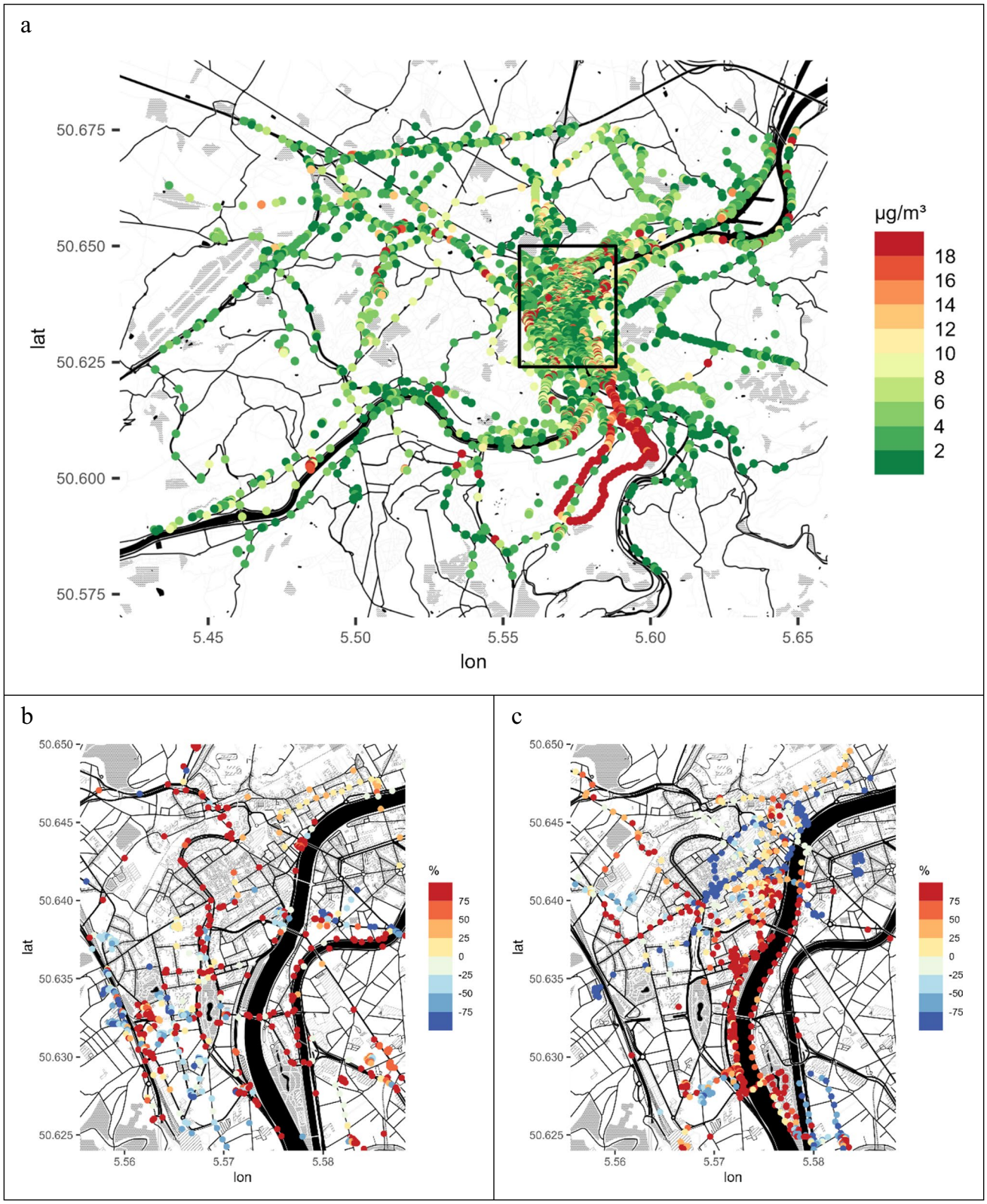

Fig. 4 Measured and modelled $PM_{2.5}$ concentrations aggregated over the all measurement periods at the model 10-m resolution. **a** Concentrations measured in the Liège city area during travels and outdoor activities (black rectangle correspond to **a** and **b** zoom area). Relative difference between modelled and measured concentrations for **b** car and **c** walk trips. Negative (blue) values indicate model underestimation; positive (red) values mean model overestimation

Journal : Large 11869	Article No : 1529	Pages : 12	MS Code : 1529	Dispatch : 31-1-2024

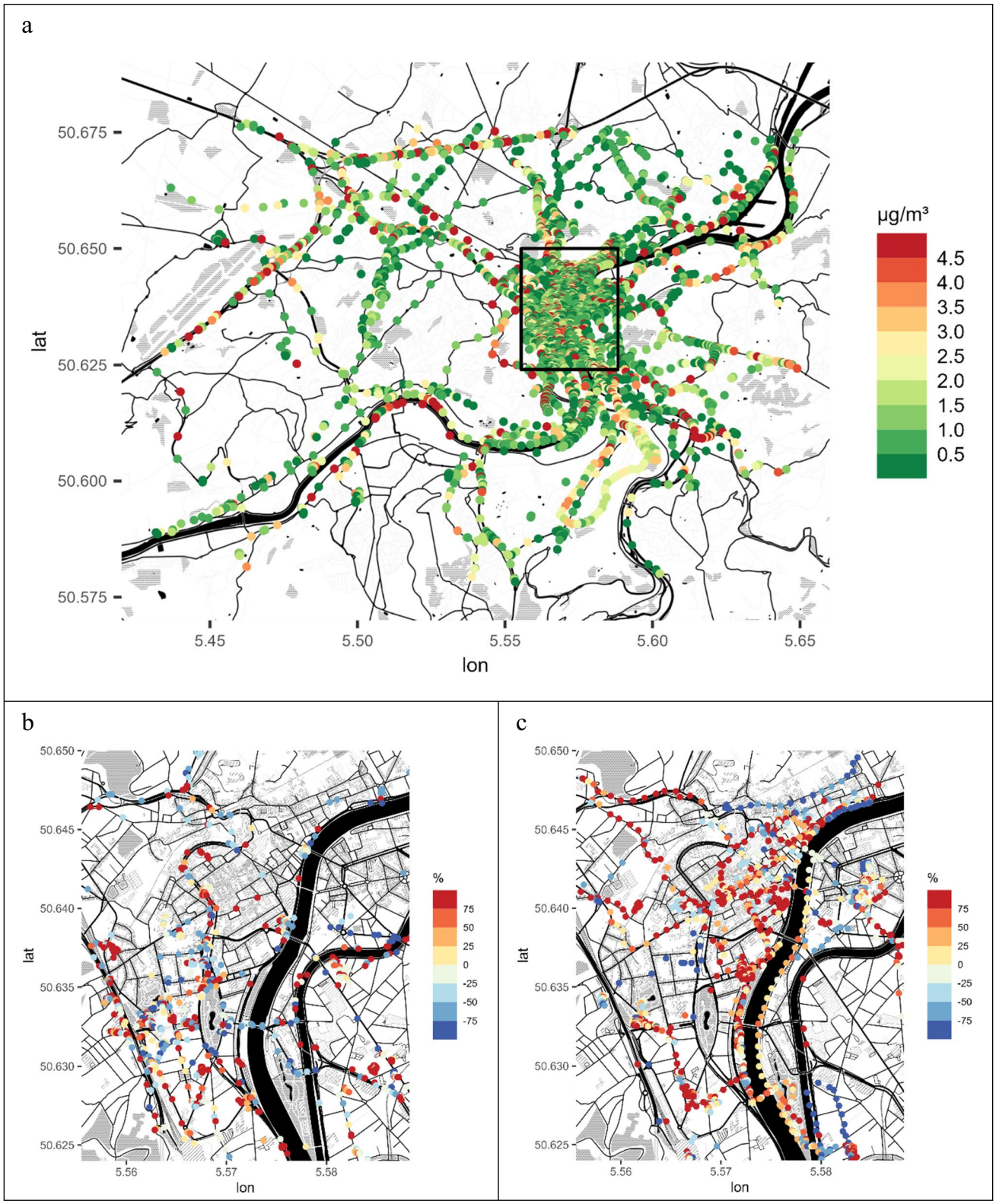

Fig. 5 Measured and modelled BC concentrations aggregated over the all measurement periods at the model 10-m resolution. **a** Concentrations measured in the Liège city area during travels and outdoor activities (black rectangle correspond to **a** and **b** zoom area). Relative difference between modelled and measured concentrations for **b** car and **c** walk trips. Negative (blue) values indicate model underestimation; positive (red) values mean model overestimation

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most exposed and the walkers and cyclists are the less ones (it should be noted that more air pollution may be inhaled as a result of accelerated and deeper breathing during physical activity). This is in agreement with the results of the ExTra-Car project (https://www.issep.be/extracar/), where home-to-office travels were made simultaneously using different modes of transportation and where bus commuters were systematically exposed to the highest levels of pollution. This citizen campaign also highlights the impacts that may have some activities with a lower frequency (and therefore less well characterized in the measurements) on personal exposure, such as indoor sport or picking up children from school for exposure respectively to $PM_{2.5}$ and BC.

The comparison of model output and measurements is far from straightforward. For example, our portable device time resolution of 1 min, adapted to track the increase and decrease of typical processes in a fix reference frame, is not ideal in a moving reference frame. The induced uncertainty concerning the position of the citizens increases with the average speed of each travel mode. The concentrations averaged at a 1-min rate might thus not be representative of the location indicated by the GPS by minute.

For the present paper, we decided to only study particulate matter ($PM_{2.5}$ and BC) because the optical sensor for the $PM_{2.5}$ measurement and BC aethalometer has been validated with success with reference analysers (Lenartz et al. 2021; Hofman et al. 2018). Measurement of particulate matter has the advantage of being less sensitive to environmental conditions than electrochemical sensors. Anyway, it remains tricky to read the highest values recorded during mobile measurements (Fig. 3). Since it is impossible to determine a posteriori if they were caused by a very high local pollution source or if they were outliers, especially when peaks were present simultaneously in both the $PM_{2.5}$ and BC time series, we kept most of them in the dataset. The regularity and accuracy of participants in keeping their activity logbook are AQ12 also critical. For activities with short duration like some car trips, precisely reporting the exact start and end of the travel is very important. For example, car trips are often characterized by a peak in BC concentrations that can be missed and associated to another activity if the logbook has been incorrectly filled. Finally, unlike systematic air pollution mapping (Apte et al. 2017) or repeated measurements performed along identical routes (Int Panis et al. 2010; Peters et al. 2014), mobile measurements of personal exposure during their daily activities with often random routes prevent to have sufficient measurement repetitions for having consistent spatial patterns over a wide area like a city. Moreover, our campaign did not cover a full year and we did not consider the meteorological conditions in the analysis yet.

The aim of this study was also to compare the mobile measurements performed during outdoor activities with the outputs of the ATMO-Street air quality model. This kind of model, which simulates pollution concentrations at the street level, is commonly used to evaluate the exposure of static population to atmospheric pollution. If measurement and model globally agree on the ranking of the level of exposure of travel modes, two main elements make the comparison difficult or even impossible: the uncertainties in individual position and the limitation of model and its inputs. The concentration measurements recorded at a fine spatiotemporal resolution were aggregated to the hourly 10-m resolution of model outputs with sometimes, as already discussed, a 1-min GPS location which does not correspond to the various environmental conditions covered by minute. Aside from inaccurate position, for car drivers and bus commuters, the concentrations were measured inside the cars (but with no information on ventilation) while the model calculates the concentrations at the façade of the buildings besides the road and not the on-the-road concentrations. Yet, the near-road horizontal concentration gradients are sharp (Sharma et al. 2009; Apte et al. 2017): NO_2 concentrations and particulate matter concentrations can decrease 30–50% within the first 250 m from the traffic emission source while BC concentrations (and NO concentrations) decrease at an even steeper rate in some cases of more than 50% within the first 200 m from the traffic emission source (Vandeninden et al. 2021). Vandeninden et al. (2021) found measured BC values on average higher than the modelled BC values for streets with a considerable amount of traffic. Here, we did not observe such underestimation of on-the-road concentrations. The comparison becomes very complicated if we try to compare measurement and model for outdoor activities such as shopping in the city during which people enter in some stores with other $PM_{2.5}$ and BC sources than traffic. The model limitations are notably linked to its input data. The discrepancies between measured and modelled concentrations can also be explained by the uncertainties in the traffic estimations and therefore in its pollutant emissions. We used the mean daily traffic derived from HERE floating car data for the year 2018. The error in the traffic estimation may be large owing to the low penetration rate of this kind of data (7.62% for the highways and 4.04% for the secondary network) and the fact that traffic dynamics like congestion are not taken into account. Finally, the meteorological conditions and background concentrations are determined using only one point of reference, which may be not enough for the studied area.

Conclusions

A quite unique amount of atmospheric pollutant measurements has been collected and contextualized in a logbook by volunteer citizens during their daily outdoor and indoor activities in Liège (Belgium). This dataset can certainly

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Journal : Large 11869 | Article No : 1529 | Pages : 12 | MS Code : 1529 | Dispatch : 31-1-2024

contribute to the assessment of personal exposure to air pollution with some interesting results presented here: (i) workers are generally less exposed during working hours than at home, (ii) the highest $PM_{2.5}$ concentrations are recorded during indoor activities and (iii) bus commuters are exposed to the highest BC levels whereas pedestrians and cyclists to the lowest ones. This study is also an original attempt to validate a street-level model with measurements during outdoor activities sometimes far away from road. The comparison of mobile measurements and ATMO-Street outputs shows that the model estimates an outdoor exposure lower than the one derived from mobile measurements. The results presented in this paper also open prospects for future research studies like leading a similar analysis for NO_2, exploiting the newly installed stationary network of low-cost sensor systems in Liège, simulating the outdoor-indoor pollutant transfer in cars (Snifecar project, https://www.issep.be/snifecar/) and launch a new project in citizen science and modelling focusing on indoor air quality only.

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AQ13 **Author contribution** Marie Dury and Fabian Lenartz contributed to the study conception and design. Material preparation, data collection and analysis were performed by Marie Dury, Florent Hozay and Fabian Lenartz. Model set-up and output analysis were carried out by Marie Dury and Hans Hooyberghs. The first draft of the manuscript was written by Marie Dury. All authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.

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Data availability The datasets generated during and/or analysed during the current study are available from the corresponding author on reasonable request.

Declarations

Consent to participate Informed consent was obtained from all individual participants included in the study.

Competing interests The authors no competing interests.

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