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Environment International 158 (2022) 106990

Contents lists available at ScienceDirect

Environment International

journal homepage: www.elsevier.com/locate/envint

Personal exposure to concentrations and inhalation of black carbon according to transport mode use: The MobiliSense sensor-based study

Sanjeev Bista *, Clélie Dureau, Basile Chaix

Sorbonne Université, INSERM, Institut Pierre Louis d'Epidémiologie et de Santé Publique IPLESP, Nemesis team, Faculté de Médecine Saint-Antoine, 27 rue Chaligny, 75012 Paris, France

ARTICLE INFO

Handling Editor: Hefa Cheng

Keywords:
Black carbon
Personal exposure
Inhaled dose
Transport pollution
Ventilation rate
Accelerometer

ABSTRACT

Introduction: Epidemiological evidence suggests that motorized vehicle users have a higher air pollutant exposure (especially from vehicle exhaust) than active (walking or cycling) transport users. However, studies often relied on insufficiently diverse sample and ignored that minute ventilation has an effect on individuals' inhaled dose. This study examined commuters' breathing zone concentration and inhaled doses of black carbon (BC) when travelling by different transport modes in the Grand Paris region.

Methods: Personal exposure to BC was continuously measured with MicroAethalometer (MicroAeth AE51) portable monitors strapped on participants' shoulder with tube inlet at the level of the neck (breathing zone), and inhaled doses were derived from several methods estimating ventilation [based on metabolic equivalents from accelerometry [METs], heart rate, and breathing rate]. Trip stages and transport modes were assessed from GPS and mobility survey data. Breathing zone concentrations and inhaled doses of BC were compared across transport modes at the trip stage level (n = 7495 for 283 participants) using linear mixed effect models with a random intercept at individual level.

Results: Trip stages involving public transport and private motorized transport were associated with a 2.20 μg/m^3 (95% CI: 1.99, 2.41) and 2.29 μg/m^3 (95% CI: 2.10, 2.48) higher breathing zone concentration to BC than walking, respectively. Trip stages with other active modes had a 0.41 μg (95% CI: 0.25, 0.57) higher inhaled dose, while those involving public transport and private motorized transport had a 0.25 μg (95% CI: −0.35, −0.15) and 0.19 μg (95 %CI: −0.28, −0.10) lower inhaled dose of BC per 30 min than walking.

Conclusion: The ranking of transport modes in terms of personal exposure was markedly different when breathing zone concentrations and inhaled doses were considered. Future studies should take both into account to explore the relationship of air pollutants in transport microenvironments with physiological response.

1. Introduction

Black carbon (BC), generated from incomplete combustion of fossil fuels (Bond et al., 2013), and several other car traffic emissions correlated with it (e.g., nitrogen dioxide, carbon monoxide) have been found to provoke chronic health problems like adverse respiratory outcomes (Lin et al., 2011) and an impairment of lung function (Suglia et al., 2008 Oct), neurological issues (Power et al., 2011 May), or cardiovascular diseases (Gan et al., 2011 Apr). A systematic review and several individual studies suggested that BC, a crucial component of particulate matters and a good indicator to classify the exposure to diesel exhaust (HEI, 2010), is one of the most valuable indicators to study traffic-related health effects (Mordukhovich et al., 2009; Liu et al., 2009; Schwartz, 2005; Diaz Resquin et al., 2018; Janssen et al., 2011).

Generally, epidemiological studies assign exposure to air pollutants at the place of residence estimated from one or several nearby air monitoring stations or from air dispersion models (Fuks et al., 2011; Jacquemin et al., 2013). This method neglects the spatial and temporal variations in exposure for each person (due to mobility and other life circumstances), which is far higher than the variations captured at fixed stations (Nieuwenhuijsen et al., 2015). Instead, continuous measurement of person level exposure with a portable device is helpful to reduce exposure misclassification (Nieuwenhuijsen et al., 2015; Setton et al., 2011), especially when studying in-vehicle exposure or exposures related to transport behaviour which are highly varying in space and time (Strickland et al., 2011).

* Corresponding author at: UMR-S 1136, Faculté de Médecine Saint-Antoine, 27 rue Chaligny, 75012 Paris, France.
E-mail address: sanjeev.bista@iplesp.upmc.fr (S. Bista).

https://doi.org/10.1016/j.envint.2021.106990
Received 23 February 2021; Received in revised form 19 October 2021; Accepted 16 November 2021
Available online 20 November 2021

The level of air pollutants to which people are exposed while travelling is higher than the benchmarks set by air quality guidelines (Morales Betancourt et al., 2017 May). It has been emphasized that although very small portions of peoples' daily schedule are spent in commuting, travelling accounts for a larger share of their daily total personal exposure as well as inhaled doses (Dons et al., 2011; Rivas et al., 2016; Beckx et al., 2009). In a Flemish population, travel periods accounting for 6.3% of subjects' daily time were shown to produce 21% of personal exposure and 29.8% of BC inhaled dose (Dons et al., 2012). Likewise, in Paris, 5.2% of daily time of students spent in travelling was responsible for 12.5% of 24-hour total BC exposure (Paunescu et al., 2017).

A number of studies have investigated the relationship between transport mode and personally measured concentrations and inhaled doses of BC. Such studies have been conducted in several cities, i.e. in Barcelona (Moreno et al., 2015; de Nazelle et al., 2012); in Lisbon (Correia et al., 2020); in Shanghai (Li et al., 2015); in London (Rivas et al., 2017); in Toronto (Jeong et al., 2017); in Singapore (Tan et al., 2017; Tran et al., 2020); in Hong Kong (Chen et al., 2020); in Bogota (Morales Betancourt et al., 2017; Morales Betancourt et al., 2019); in Stockholm (Merritt et al., 2019); in California (Ham et al., 2017); in Londrina (Targino et al., 2018); and in Mexico City (Velasco et al., 2019). In 2017; a systematic review including 39 papers concluded that motorized vehicle users are facing higher levels of air pollution exposure compared to pedestrians (Cepeda et al., 2017); whereas some individual studies reported the opposite (de Nazelle et al., 2012; Briggs et al., 2008; Quiros et al., 2013).

However, studies have often ignored ventilation rates when quantifying transport-related air pollution exposure. Such studies assume a uniform breathing rate and neglect that the amount of pollutants inhaled (going to lungs) is dependent on the breathing rate (Dons et al., 2017). Considering inhaled doses over personally measured concentrations is useful to overcome measurement error and improves accuracy while estimating transport-related health risks (Cepeda et al., 2017). This is particularly true when the aim is to establish health benefits of cycling and walking, as the higher inhaled dose of pollutants may offset some of the health benefits gained through physical activity (Laeremans et al., 2018; Giles and Koehle, 2014).

Of the work referred to above dedicated to BC by transport modes, studies either did not considered inhaled doses (Rivas et al., 2017; Jeong et al., 2017; Merritt et al., 2019; Ham et al., 2017), or involved only one method for the estimation of inhaled doses (de Nazelle et al., 2012; Correia et al., 2020; Li et al., 2015; Tan et al., 2017; Morales Betancourt et al., 2019; Targino et al., 2018), or were experimental studies not considering a large number of different trips (de Nazelle et al., 2012; Correia et al., 2020; Rivas et al., 2017; Jeong et al., 2017; Tan et al., 2017; Morales Betancourt et al., 2019; Ham et al., 2017; Targino et al., 2018 27), or had a small sample size of trips (less than 500 or even 200) (de Nazelle et al., 2012; Correia et al., 2020; Li et al., 2015; Rivas et al., 2017; Jeong et al., 2017; Tan et al., 2017; Morales Betancourt et al., 2019; Targino et al., 2018; Velasco et al., 2019), or only considered specific hours of the day or specific days of the week or specific seasons (de Nazelle et al., 2012; Correia et al., 2020; Li et al., 2015; Rivas et al., 2017; Jeong et al., 2017; Tan et al., 2017; Morales Betancourt et al., 2019; Merritt et al., 2019; Targino et al., 2018), or did not consider walking as a separate mode (Correia et al., 2020; Rivas et al., 2017; Ham et al., 2017).

The first aim of this study is to analyse the relationship between personal use of transport modes and personally measured concentrations of BC in the breathing zone, at the level of trip stages. The second aim is to compare differences of exposure to BC by transport modes when exposure is defined as personal breathing zone concentration versus inhaled doses.

2. Material and method

2.1. Study design

This study was conducted in the Grand Paris (the Paris City and a number of surrounding municipalities) in France from May 2018 to October 2020 (first wave of the MobiliSense project funded by the European Research Council). Participants were recruited following a two-stage stratified random sampling procedure. Stage 1 pertained to the random selection of neighbourhoods in the first and last quartiles of road traffic density in each quartile of area income. The second stage involved the random selection of dwelling units in the pre-selected neighborhoods from the 2013 and 2014 population censuses by the National Institute of Statistics and Economic Studies. Overall, 31970 dwellings were selected from 234 neighborhoods. Postal mails were sent twice to invite residents from the selected dwellings to participate. Overall, 289 participated in the sensor-based MobiliSense study. An analysis reported in Appendix 1 shows that older participants, people with higher education, inactive people and to a lower extent self-employed people and people with stable and unstable jobs (compared to unemployed people), and French citizens had a higher probability of participating in our study.

2.2. Data collection and initial processing

Participants were followed for 4 continuous days from day 1 (3 am) to day 5 (3 am) with a MicroAethalometer (MicroAeth AE51, AethLabs, CA, USA). Personally measured concentrations of BC around the breathing zone was continuously measured at a 10 s time resolution with the AE51 strapped on participants' shoulder with the inlet of the tube at the level of neck. AE51 is a valid device (Cai et al., 2014) that has already been used in multiple studies (Morales Betancourt et al., 2017; de Nazelle et al., 2012; Apte et al., 2011; Cheng and Lin, 2013). This pocket-sized device analyses BC mass by examining the changes in absorption of transmitted light [coefficient of light attenuation (ATN)] at 880 nm on a filter strip housing a small-sized Teflon-coated borosilicate glass fibre filter (T60). BC concentration measured by AE51 can be substantially underestimated when the filter is overloaded. Thus, to avoid filter saturation and preserve measurement integrity, participants were asked to change the filter on the second day at 8 pm when they were recruited in winter, and the filter was changed in any case by a new one whenever the device indicated necessity of filter replacement (when the optical density reached a certain level). Therefore, data do not need to be corrected for any sort of filter loading effects (Correia et al., 2020). The post processing of BC data was done using the Optimized Noise Reduction Averaging (ONA) algorithm (Hagler et al., 2011) (see details of the ONA algorithm in Appendix 2). We provide our version of the algorithm in the R package "BlackCarbon" version 1.1 developed by Bista 2020 (BlackCarbon, 2020). The ONA algorithm has to take into account filter changes (Hagler et al., 2011). Filter changes were identified from a ΔATN of at least -5 between two successive data points, and these pre-identified candidate for a filter change were manually verified.

Electronic or instrumental optical noise results into no change of ATN coefficient or even drop of ATN for some successive observations, as opposed to the general concept of the device (Hagler et al., 2011). This results in flawed negative concentrations or constant concentrations of BC for a relatively long period of time even after applying the ONA algorithm. Therefore, the ONA processed BC concentrations that remained negative for longer than 5 min or trip stages fully overlapping time periods having a constant concentration of BC for more than 4 h, although the person was switching between micro-environments, were deleted from the analysis.

Heart rate and breathing rate data were continuously collected on the 2nd and 4th days by a portable Zephyr BioHarness 3.0 device (Nazari et al., 2018) at 1 s time intervals. The heart rate and breathing rate data

processing followed steps suggested by Bigazzi and Figliozzi (Bigazzi and Figliozzi, 2015).

ActiGraph wGT3X + devices (Corporation, 2013) worn by participants on their waist throughout the study period collected accelerometer data in 1 s epochs. ActiGraph devices have been shown to be reliable in detecting different levels of physical activity (Metcalf et al., 2002). In consideration of people who move slowly, the low-frequency extension option was chosen over normal filter in Actilife 6.13.3 (Wanner, 2013). A 3-axes count of 0 for at least 1 h with a spike tolerance of 2 min of non-zero epochs, a default setting in Actilife, was used to identify non-wear time of the accelerometer (Troiano et al., 2008; Chaix et al., 2014). The refined 2-regression model proposed by Crouter (Crouter et al., 2010) was used in Actilife to calculate the Metabolic Equivalents (METs) at 10 s epochs.

Participants indicated non-wear time of each device used in the study, which was confirmed with participants via personal phone calls. Confirmed non-wear time periods were deleted for each sensor and analysis periods before the final processing.

2.3. Breathing zone concentration

Personally measured breathing zone concentration of BC for each trip stage is the mean BC concentration measured with AE51 in the participants' breathing zone over the period of the corresponding trip stage. The cumulated personal exposure to breathing zone concentration of BC was calculated by multiplying the mean personal breathing zone concentration of the corresponding trip stage by the respective trip duration. Summation of these products by transport modes yielded the cumulated personal exposure to concentrations of BC by transport modes. This cumulated personal exposure was only computed to determine the percentage of exposure to concentrations by transport modes, and to compare it with the percentage of inhaled doses by transport modes.

2.4. Calculation of minute ventilation and inhaled doses

Dons et al in 2017 published a paper comparing several methods for estimating minute ventilation and made recommendations for using those based on the objective, duration, and the size of the study (Dons et al., 2017). Following the paper, different methods appropriate to our data and study type were chosen to estimate minute ventilation as specified in Appendix 3 (Appendix Table A2).

Since accelerometer data were collected throughout the study, they allowed us to include a larger number of trips stages with minute ventilation than when breathing rate and heat rate measurements recorded only on the 2nd and 4th days were used. Therefore, method 1.1, using METs (Appendix Table A2), was chosen to report inhaled doses in the main text. This technique computes the ventilation rate as the product of METs with the energy conversion factor, which is derived stochastically as a function of individual's gender and age (Appendix Table A3 in Appendix 3).

After merging BC data with ventilation data to the nearest 5 s data point, inhaled doses were calculated at a 10 s resolution as the product of the corresponding breathing zone concentration and minute ventilation using the following equation (Apparicio et al., 2018):

$$\text{Inhaled dose} = (\text{VE}*0.001) * \text{concentration of BC}$$

where the inhalation is in μg/minute, VE is the ventilation rate in liter/minute, and the concentration is in $\mu g/m^3$.

2.5. Classification of trip stages

Trip stages are segments of trips with a unique transport mode used. Successive trip stages within a trip are necessarily separated by a time segment of mode transfer assigned to a stationary location.

After completion of the sensor data collection, data extracted from the BT-Q1000XT GPS receiver were uploaded in the TripBuilder web mapping application (Oliveira et al., 2011; Wolf et al., 2004) that pre-processed them to identify the start and end times of each trip stage and related transport modes (see detail in Appendix 4). Using the TripBuilder application, the trip stage data over the whole period (including trip stage start and end and mode) were verified with the participants during a phone mobility survey. The final output is a timetable including time-stamped information on trip stages and related information.

A crude classification of transport modes at the trip stage level included the following: entirely walking, other active mode (skateboard, roller skate, biking, etc.), personalized motorized vehicle (including taxi and motorbike), and public transport. A more detailed classification included entirely walking, other active mode, driver of personal motorized vehicle, passenger of personal motorized vehicle (including taxi), bus/coach, tramway, metro (within Paris city), and suburban trains.

Reasons for excluding trips from the present analysis (summarized in Fig. 1) included non-wear of either the AE51 or the wGT3X+, lack of data due to recording error, trips made by long distance transport modes or jogging trips (which are not classical daily transport modes or could induce vibrations), etc. Long distance trip in our study is defined as a trip made by airplane or long distance train (distinct from suburban trains). The average duration of long distance trips in our study was 1 h and 45 min (350 km in distance), whereas the average travel duration of other daily travel trips made in the Grand Paris region was 12 min (3.5 km in distance). Furthermore and most importantly, there were 70 trips made by long distance transport modes (airplane and long distance train) of which only 64 had BC concentration and accelerometer data. The two reasons why we excluded trips with long distance modes (plane, very high speed train) is first that these trips were markedly different than the others and second (most importantly) that the sample size for these trips was low. Overall, 7495 trip stages made by 283 participants were retained for analysis.

2.6. Background concentration of air pollutants

Background concentration of particular matter with an aerodynamic diameter of 2.5 μm or smaller ($PM_{2.5}$) and nitrogen dioxide (NO_2) were retrieved for all the geographic coordinates continuously recorded with GPS receivers and the mobility survey (as explained in Appendix 5) by matching their timestamps to those of the closest Airparif air quality monitoring stations. To minimize the misclassification in personal exposure to background concentrations of air pollutants, we ignored those points which were more than 15 km away from the nearest station. These $PM_{2.5}$ and NO_2 background concentrations at the level of mobility points were aggregated at the trip stage level using the start and end times of the trip stages. For the trip stages without continuous mobility points (n = 121 which were often short trips), we took the trips' extremities (start and end point), retrieved the $PM_{2.5}$ and NO_2 concentration from the nearest monitoring station at those points and corresponding timestamps, and determined the trip stage background concentration as the average of the start and end points concentrations. At the end, 264 trip stages had no information on $PM_{2.5}$ and 188 trip stages were lacking NO_2 background concentrations, due to the fact that they were >15 km away from the nearest monitoring station. To capture the non-linear association between background concentrations and personal BC exposure, we grouped trip stages' background concentrations into quartiles and labelled "missing" to the trip stages lacking background concentration information.

2.7. Statistical analysis

Variations of BC breathing zone concentrations and inhaled doses across transport modes at the trip stage level were examined using a

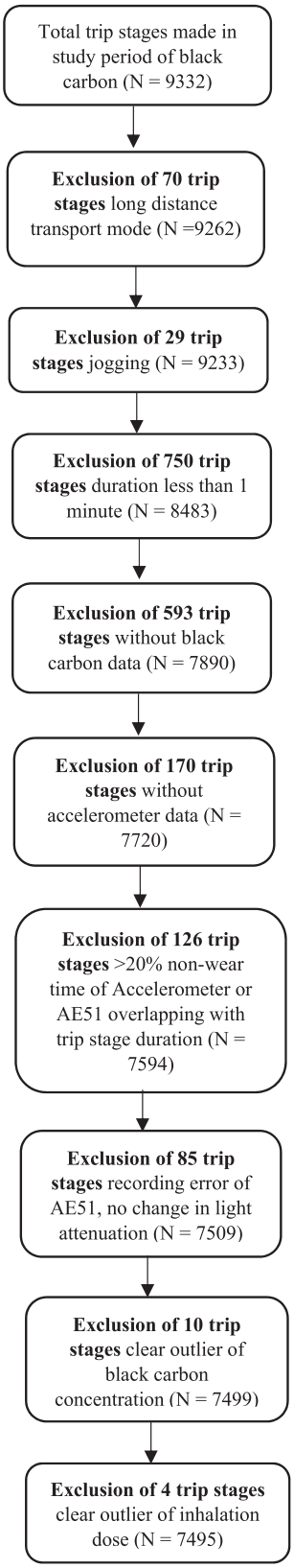

Fig 1. Data collection work flow.

linear multilevel model including a random intercept at the individual level. Observations were weighted according to trip stage durations.

Trips made in central Paris, especially by walking and biking, may have different level of BC exposure than trips made in suburbs, because of spatial variation in background BC concentration caused by differences in traffic density and proximity to green spaces. Furthermore, trips made in different seasons, days of the week, and time of the day may have different levels of BC exposure, although the trips were made via the same route and transport mode. This might be due to temporal variability in background BC concentration caused by different factors such as variation in meteorological conditions and traffic density. Therefore, in order to estimate the effect of the individual transport mode per se on the personal exposure independent of external conditions, models in our study were adjusted for temporal and spatial effects, namely: season; time of the day as morning (6 am to 11 am), afternoon (12 am to 5 pm), evening (6 pm to 9 pm) and night (10 pm to 5 am); weekday vs. weekend; and background $PM_{2.5}$ and NO_2 concentrations. Adjusting for background $PM_{2.5}$ concentration controls for the background BC concentration as BC is a major contributor to the $PM_{2.5}$ in the atmosphere (Santoso et al., 2013 Sep 29) and NO_2 is taken into account as a proxy for vehicle density as it is a good measure of vehicle emission (Carslaw et al., 2016), the major source of BC. By adjusting for these temporal and spatial effects in a regression analysis, we control for the conditions of the trips in order to estimate the effect of the individual transport mode per se on the personal exposure independent of external condition.

In the present analysis, we considered that sociodemographic characteristics should not be associated with the particular exposure during trips conditional on the chosen transport mode. Also, controlling for sociodemographic characteristics would yield an abstract estimate considering a similar sociodemographic structure across transport modes, which is not realistic.

3. Results

3.1. Descriptive data on participants and trip stages

In the sample of 283 participants, mean age was 50 years [interdecile range (IR): 38, 62 years]. Fifty-seven percent of them were female; 24% were living in Paris and the rest in the suburbs; 66% had a permanent job, 5% an unstable job, 3% were unemployed, and 12% retired; 67 % of participants had 3 or more years of University education, whereas 5% had primary or secondary level educational attainment.

Over the study period (1st to 4th day), 97% of the 283 participants (n = 276) had all 4 days of data, and 1 had 3.5 days, 2 had 2 days, and 4 only had 1 days of data. Among trip stages retained for analysis, 58.3% were entirely walked; 5.4% were with another active mode (biking/rollers/skateboard); 15.8% were with public transport and 20.5% with private motorized vehicle (Table 1). Participants had a median number of trip stages of 6 (IR: 3, 10) and the median time spent on travelling by a person per day was 1 h and 26 min (IR: 37 min, 2 h 18 min). The detail by transport mode is reported in Table 1.

3.2. Breathing zone concentrations and inhaled doses of BC by transport modes

Participants were exposed to a median breathing zone concentration of 1.82 μg/m^3 of BC during transport activity (IR: 0.80, 4.09) while they were exposed to a median concentration of 0.95 μg/m^3 (IR: 0.46, 1.49) during non-transport time (in the distribution of 283 participants). Participants inhaled more than twice the dose of BC while they were travelling [median: 0.89 μg per 30 min (IR: 0.43, 1.84)] compared to non-transport time [median: 0.35 μg per 30 min (IR: 0.16, 0.64)]. Participants inhaled a median of 3.11 μg of BC per day during transport (IR: 1.10, 7.08).

Table 2 shows descriptive statistics on BC breathing zone concentrations, inhaled doses, and minute ventilation weighted by trip duration for the different transport modes considered, while Figs. 2–4 show the corresponding density plots. While active modes (walking and other

Table 1
Descriptive statistics on trip stages according to the transport mode used.

Classifications of trip stages according to the mode	Number of trip stages (%)	Number of trip stages per participant per day[a] Median (10th and 90th percentiles)	Duration of trip stages (in minutes) per participant per day[a] Median (10th and 90th percentiles)
Crude classification			
Entirely walked trips	4372 (58.3)	3.7 (1.0, 7.0)	28.5 (4.2, 67.5)
Other active modes	407 (5.4)	0.0 (0.0, 1.5)*	0.0 (0.0, 21.9)*
Public transport	1183 (15.8)	0.5 (0.0, 3.0)*	4.2 (0.0, 47.0)*
Private motorized	1533 (20.5)	1.0 (0.0, 3.5)*	17.9 (0.0, 68.6)*
P-value (Kruskal-Wallis test)	NA	<0.001	<0.001
Detailed classification			
Entirely walked trips	4372 (58.3)	3.7 (1.0, 7.0)	28.5 (4.2, 67.5)
Other active modes	407 (5.4)	0.0 (0.0, 1.5)*	0.0 (0.0, 21.9)*
Bus/coach	210 (2.8)	0.00 (0.0, 0.7)*	0.0 (0.0, 8.3)*
Metro	608 (8.1)	0.0 (0.0, 1.7)*	0.0 (0.0, 24.8)*
Suburban train	290 (3.9)	0.0 (0.0, 1.0)*	0.0 (0.0, 17.0)*
Tramway	75 (1.0)	0.0 (0.0, 0.0)*	0.0 (0.0, 0.0)*
Private motorized (driver)	1277 (17.0)	0.5 (0.0, 3.2)*	8.4 (0.0, 65.4)*
Private motorized (passenger)	256 (3.4)	0.0 (0.0, 0.7)*	0.0 (0.0, 15.1)*
P-value (Kruskal-Wallis test)	NA	<0.001	<0.001

MobiliSense Study, 283 participants, 7495 trip stages.
*Statistically significant at 0.05 alpha level compared to entirely walked trip stages (conclusion drawn from Bonferroni adjusted p-values from Wilcoxon tests).
NA: Not applicable.
[a] The numbers and durations of trip stages are calculated across the 283 individuals, including those who do not use the corresponding modes.

active modes) were associated with lower breathing zone concentration than motorized modes, they were not related to lower inhaled doses, which is as shown in the last two columns explained by the much higher minute ventilation in such active modes. The highest inhaled dose was documented in other active mode trip stages, with a median of 1.45 μg per 30 min. When the detailed classification of trip stages was examined, metro trip stages were also associated with a high median inhaled dose, of 1.40 μg per 30 min, which is related to the fact that the median BC breathing zone concentration was almost twice higher for metro trip stages, 4.83 μg/m^3, than for other active modes, 2.34 μg/m^3.

To compare the distribution of breathing zone concentrations and inhaled doses, we calculated a cumulated personal exposure in μg/m^3 corresponding to a summation over time of the breathing zone concentration. Of the cumulated transport-related exposure to BC breathing zone concentration (Table 3), the highest fraction, 41.8%, was related to private motorized trip stages followed by public transport and entirely walked trip stages corresponding to 26.1% and 25.5% of the exposure respectively. On the opposite, entirely walked trip stages accounted for a much larger portion of cumulated transport-related BC inhaled doses, i.e., 43.2%, followed by private motorized trip stages corresponding to 28.5%.

3.3. Results from multilevel models

Associations estimated from multilevel models are reported in Table 4. Regarding breathing zone concentrations of BC, using public transport (+2.20 μg/m^3, 95% CI: 1.99, 2.41) but also personal motorized vehicles (+2.29 μg/m^3, 95% CI: 2.10, 2.48) was associated with a higher average breathing zone concentration compared to walking. When using the detailed classification of modes, metro trip stages were found to be associated with the highest breathing zone concentration (3.08 μg/m^3, 95% CI: 2.82, 3.34) compared to walking. It was also found that drivers of private motorized vehicles were exposed to higher BC concentrations than passengers.

As shown in Table 4, breathing zone concentrations and inhaled doses of BC did not lead to the same ranking of transport modes, due to the varying minute ventilation observed across modes. Other active modes were still associated with a 0.41 μg (95% CI: 0.25, 0.57) higher inhaled dose of BC per 30 min compared to walked trip stages. While they were associated with a higher breathing zone concentration, private motorized trip stages were related to a 0.19 μg (95% CI: 0.28, 0.10) lower inhaled dose of BC per 30 min than walked trip stages, due to their lower minute ventilation.

Similarly, while public transport trip stages were related to higher BC breathing zone concentrations than walked trip stages, they were associated with a 0.25 μg (95% CI: 0.35, 0.15) per 30 min lower inhaled dose overall. However, when the detailed classification of transport modes

Table 2
Overall breathing zone concentration and inhaled dose of black carbon in trip stages according to the transport mode used.

Classifications of trip stages according to the mode	Breathing zone concentration of black carbon (μg/m^3) at the trip stage level weighted by duration of trip stages		Inhaled dose of black carbon (μg/ 30 min) at the trip stage level weighted by duration of trip stages		Minute ventilation (liter/minute) at the trip stage level weighted by duration of trip stages	
	Median (10th and 90th percentiles)	Mean (standard deviation)	Median (10th and 90th percentiles)	Mean (standard deviation)	Median (10th and 90th percentiles)	Mean (standard deviation)
Crude classification						
Entirely walked trips	1.35 (0.35, 4.18)	2.04 (2.63)	0.98 (0.21, 3.03)	1.43 (1.64)	24.89 (13.63, 33.26)	24.31 (7.47)
Other active modes	2.34 (0.70, 5.46)*	3.01 (3.24)	1.45 (0.44, 3.52)*	1.90 (2.46)	20.52 (13.38, 27.53)*	20.70 (5.73)
Public transport	3.84 (0.90, 9.23)*	4.56 (3.56)	1.10 (0.25, 2.68)*	1.33 (1.10)	10.30 (6.30, 16.99)*	11.05 (4.51)
Private motorized	3.45 (0.62, 9.15)*	4.36 (3.97)	0.91 (0.18, 2.69)*	1.23 (1.21)	9.18 (6.76, 13.70)*	9.90 (3.20)
P-value (Kruskal-Wallis test)	<0.001		<0.001		<0.001	
Detailed classification						
Entirely walked trips	1.35 (0.35, 4.18)	2.04 (2.63)	0.98 (0.21, 3.03)	1.43 (1.64)	24.89 (13.63, 33.26)	24.31 (7.47)
Other active modes	2.34 (0.70, 5.46)*	3.01 (3.24)	1.45 (0.44, 3.52)*	1.90 (2.46)	20.52 (13.38, 27.53)*	20.70 (5.73)
Bus/coach	2.57 (0.84, 6.13)*	3.12 (2.54)	0.78 (0.27, 2.15)	1.06 (0.96)	11.38 (6.99, 16.40)*	11.92 (4.25)
Metro	4.83 (1.42, 10.21)*	5.51 (3.68)	1.40 (0.46, 3.02)*	1.64 (1.14)	11.23 (6.87, 17.85)*	12.06 (4.65)
Suburban train	3.32 (0.67, 9.23)*	4.35 (3.56)	0.84 (0.17, 2.30)	1.12 (1.02)	8.33 (5.76, 13.65)*	9.40 (3.78)
Tramway	1.39 (0.18, 5.09)	2.36 (2.57)	0.30 (0.05, 1.50)*	0.60 (0.83)	6.37 (4.98, 11.50)*	7.79 (3.72)
Private motorized (driver)	3.54 (0.64, 9.49)*	4.49 (4.02)	0.96 (0.19, 2.78)*	1.28 (1.24)	9.17 (6.77, 13.72)*	9.93 (3.21)
Private motorized (passenger)	2.75 (0.61, 7.81)*	3.72 (3.57)	0.74 (0.16, 2.38)*	1.01 (1.02)	9.25 (6.53, 13.65)*	9.79 (3.11)
P-value (Kruskal-Wallis test)	<0.001		<0.001		<0.001	

MobiliSense Study, 283 participants, 7495 trip stages.
Note: Minute ventilation was estimated from METs and was used to calculate the inhaled dose.
*Statistically significant at 0.05 alpha level compared to entirely walked trip stages (conclusion drawn from Bonferroni adjusted p-values from Wilcoxon tests).

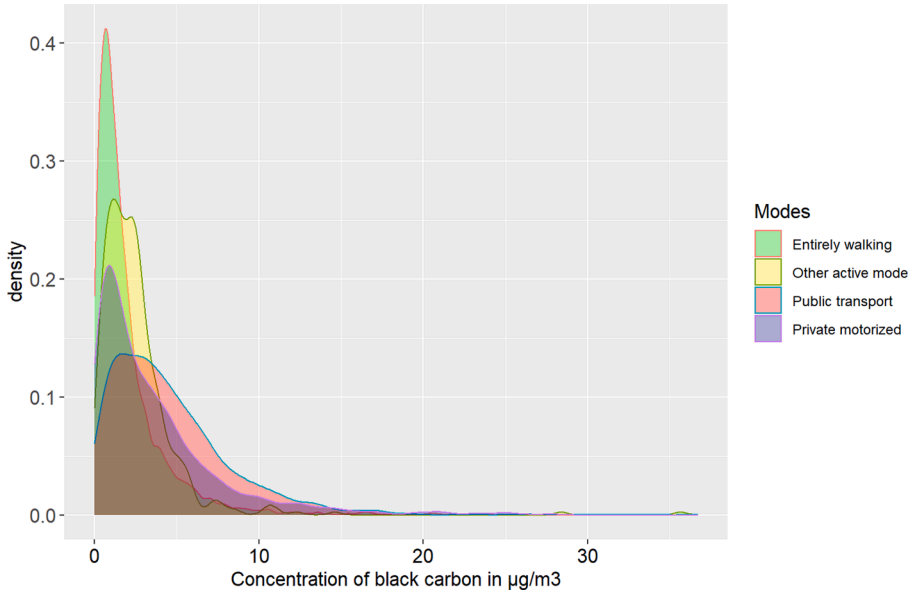

Fig 2. Density plot of black carbon concentration (µg/m^3) by mode of transportation (crude classification).

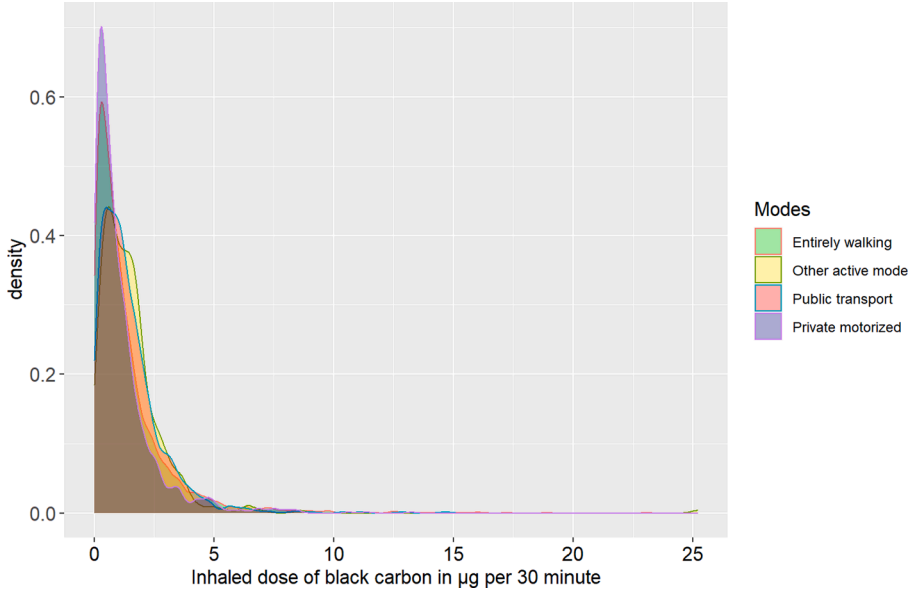

Fig 3. Density plot of per 30-minute inhaled dose of black carbon (µg) by mode of transportation (crude classification).

was analyzed, we found that metro trip stages were not different than walked trip stages in terms of inhaled dose of BC per 30 min, whereas trip stages with busses, suburban trains, and tramways implied lower inhaled doses of BC than walking.

The different methods for calculating minute ventilation are compared in Appendix 6. As shown in Appendix 7 (Appendix Table A10), there were differences in the ordering of transport modes with respect to exposure when inhaled doses were derived from different methods for calculating ventilation. However, for all the methods, the other active modes category was associated with the highest inhaled doses. Depending on the method, motorized modes were associated with a slightly lower or a slightly higher inhaled dose than walking.

4. Discussion

Using an advanced sensor-based approach to data collection and a trip stage level analytical design, this study documents that public transport (especially metro, bus, and suburban train), private motorized transport, and other active modes were associated with a higher breathing zone concentration of BC compared to transport walking. This is in line with the findings of a study done in Shanghai (Li et al., 2015), where the average BC exposure were 5.59 µg/m^3, 6.58 µg/m^3, 7.28 µg/m^3, 8.62 µg/m^3 and 9.43 µg/m^3 for walking, cycling, bus, taxi and subway trips respectively, and with several other studies (de Nazelle et al., 2012; Williams and Knibbs, 2016). In our study, particularly high breathing zone concentration of BC were documented during metro trip stages. Even if metros are not powered by combustion of fossil fuels, BC from the outer environment may enter into the metro underground space via the ventilation system which is often located on the ground of roads. BC particles have small diameters (<1 µm), which makes them hard to be efficiently blocked by the filters installed in metro vehicles (Gong et al., 2016). Moreover, to perform work on the underground railway network, the company and workers use locomotives powered by diesel engines, which might to some extent contribute to the observed

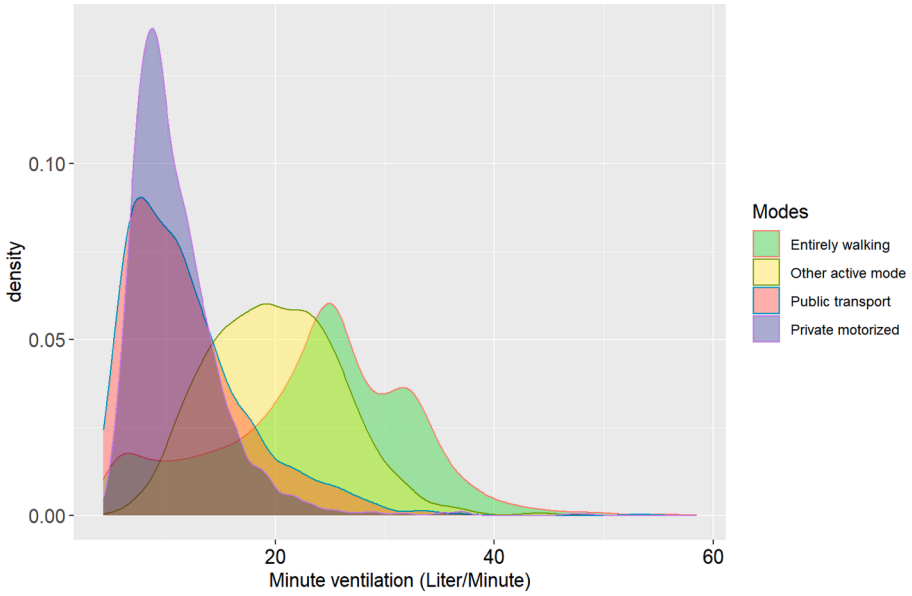

Fig 4. Density plot of minute ventilation (Liter/Minute) by mode of transportation (crude classification).

Table 3
Cumulated inhalation of black carbon in trip stages per individual per day and comparative distribution of inhaled doses and cumulated exposure to breathing zone concentrations across transport modes.

Classifications of trip stages according to the mode	**Cumulated inhalation of black carbon per individual[a] per day in µg: median (10th and 90th percentiles)**	**% of black carbon inhaled attributable to these trip stages**	**% of cumulated breathing zone concentrations of black carbon attributable to these trip stages**
Crude classification			
Entirely walked trips	1.11 (0.14, 3.82)	43.2	25.5
Other active modes	0.00 (0.00, 0.88)	10.0	6.6
Public transport	0.11 (0.00, 1.98)	18.3	26.1
Private motorized	0.40 (0.00, 3.27)	28.5	41.8
Detailed classification			
Entirely walked trips	1.11 (0.14, 3.82)	43.2	25.5
Other active modes	0.00 (0.00, 0.88)	10.0	6.6
Bus/coach	0.00 (0.00, 0.30)	2.8	3.4
Metro	0.00 (0.00, 1.36)	10.5	14.7
Suburban train	0.00 (0.00, 0.55)	4.6	7.4
Tramway	0.00 (0.00, 0.00)	0.4	0.6
Private motorized (driver)	0.19 (0.00, 3.04)	24.8	36.1
Private motorized (passenger)	0.00 (0.00, 0.47)	3.8	5.8

MobiliSense Study, 283 participants, 7495 trip stages.
Note: Minute ventilation was estimated from METs and was further used to calculate the inhaled dose.
[a] The numbers and durations of trip stages are calculated across the 283 individuals, including those who do not use the corresponding modes.

findings. Finally, primary sources of particles in the metro environment are iron particles resulting from mechanical friction between rails, wheels and brakes (Moreno et al., 2015). Since iron and BC absorb light at similar wavelength (Karanasiou et al., 2015), this may result in the overestimation of BC concentration in metros while using AE51.

The slightly higher breathing zone concentration observed for other active modes in comparison to walking may be due to the closer proximity of biking lanes than footpaths from vehicular emissions. The lowest breathing zone concentrations of BC were reported for tramway, as tramway uses a separated dedicated lane (thus relatively distant from road vehicles) to run and is operated electrically. Still, the BC concentration level documented in tramways likely comes from on-road vehicle exhaust getting into tramway due to ventilation or opening of doors.

When inhaled doses rather than concentrations were considered, other active modes (bicycle, skate boards, etc.) appeared to be associated with a higher inhaled dose of BC per 30 min than other modes. This is mirroring the conclusion of a study done in Belgium where biking was documented with an average inhaled dose of 200 ng/minute, whereas several other modes (such as train, metro, bus, and car) had BC inhaled dose lower than 100 ng/minute (Dons et al., 2012). Also, studies done in Lisbon and Barcelona documented that bicycle trips had a higher average BC inhaled dose than trips made with metro, bus, and private car (de Nazelle et al., 2012; Correia et al., 2020). We found that trip stages made by other active modes imply an increased level of physical activity (and a high rate of ventilation), which results in a higher inhalation (Dons et al., 2017; Laeremans et al., 2018; Giles and Koehle, 2014), although breathing zone concentrations of BC are comparatively lower than for several motorized modes. It suggests that dose–response relationships of air pollutant exposure in transport microenvironments with health status should be explored while taking into account inhaled doses rather than only measured concentrations.

Inconsistencies in the ordering of transport modes with respect to inhaled doses between the different methods of calculation of the ventilation rate (Appendix Table A8 in Appendix 6, Appendix Table A10 in Appendix 7) are related to the fact that the calculated minute ventilation has a substantial impact in determining the quantity of air pollutants inhaled (Dons et al., 2017). Method 1.1 used METs, age, and sex, while method 2.1–2.3 used heart rate and sex, and only 3.1 used breathing rate for the calculation. A calculation based on MET level (method 1.1) assigns lower minute ventilation to the non-physically active modes (such as suburban train, private motorized vehicles, and tram) compared to the heart rate and breathing rate methods because accelerometers detect no activity when people are usually sitting, whereas heart and breathing rates imply a weaker discontinuity in the data (Dons et al., 2017). Meanwhile, in bus and metro, people often travel standing, and some acceleration is therefore detected and thus a higher minute ventilation is assigned than with other motorized modes

Table 4
Trip stage-level associations between the transport mode used and personal breathing zone concentration and inhalation of black carbon[a].

Classifications of trip stages according to the mode	Breathing zone concentrations of black carbon (μg/m^3) β (95% CI)[+] Adjusted	Inhaled dose of black carbon (μg per 30 min of travel time) β (95% CI)[+] Adjusted	Minute ventilation (liter/minute) β (95% CI)
Crude classification (Model 1)			
Entirely walked trips	Ref	Ref	Ref
Other active modes	1.03 (0.68, 1.38)	0.41 (0.25, 0.57)	−4.76 (-5.36, −4.16)
Public transport	2.20 (1.99, 2.41)	−0.25 (-0.35, −0.15)	−13.06 (-13.41, −12.71)
Private motorized	2.29 (2.10, 2.48)	−0.19 (-0.28, −0.10)	−14.45 (-14.79, −14.11)
Seasons			
Spring	Ref	Ref	NA
Winter	0.60 (0.09, 1.11)	0.28 (0.06, 0.50)	
Autumn	0.49 (0.02, 0.96)	0.18 (-0.02, 0.38)	NA
Summer	0.75 (0.24, 1.26)	0.28 (0.06, 0.50)	NA
Time of the day			
Night	Ref	Ref	NA
Afternoon	0.00 (-0.40, 0.40)	−0.39 (-0.58, −0.20)	NA
Evening	0.08 (-0.33, 0.49)	−0.31 (-0.50, −0.12)	NA
Morning	0.60 (0.20, 1.00)	−0.11 (-0.30, 0.08)	NA
Weekday Vs weekend	0.42 (0.23, 0.61)	0.22 (0.13, 0.31)	NA
$PM_{2.5}$(μg/m^3)			
1st quantile (0.00 to 7.30)	Ref	Ref	NA
2nd quantile (7.31 to 11.23)	0.38 (0.16, 0.60)	0.14 (0.04, 0.24)	NA
3rd quantile (11.24 to 17.20)	0.46 (0.21, 0.71)	0.22 (0.11, 0.33)	NA
4th quantile (17.21 to 89.21)	1.14 (0.87, 1.41)	0.59 (0.46, 0.72)	NA
Missing (n = 264 trip stages)	0.20 (-0.48, 0.88)	0.11 (-0.21, 0.43)	NA
NO_2 (μg/m^3)			
1st quantile (0.70 to 23.20)	Ref	Ref	NA
2nd quantile (23.21 to 43.20)	0.49 (0.27, 0.71)	0.29 (0.19, 0.39)	NA
3rd quantile (43.21 to 65.60)	0.84 (0.59, 1.09)	0.35 (0.24, 0.46)	NA
4th quantile (65.61 to 383.10)	1.44 (1.16, 1.72)	0.57 (0.44, 0.70)	NA
Missing (n = 188 trip stages)	−0.48 (-1.22, 0.26)	−0.14 (-0.48, 0.20)	NA
Detailed classification (Model 2)			
Entirely walked trips	Ref	Ref	Ref
Other active modes	1.03 (0.68, 1.38)	0.41 (0.25, 0.57)	−4.76 (-5.36, −4.16)
Bus/coach	1.41 (0.99, 1.83)	−0.33 (-0.53, −0.13)	−12.68 (-13.41, −11.95)
Metro	3.08 (2.82, 3.34)	0.06 (-0.06, 0.18)	−11.80 (-12.25, −11.35)
Suburban train	1.53 (1.20, 1.86)	−0.63 (-0.79, −0.47)	−14.97 (-15.54, −14.40)
Tramway	0.01 (-0.74, 0.76)	−0.94 (-1.29, −0.59)	−15.74 (-17.03, −14.45)
Private motorized (driver)	2.31 (2.10, 2.52)	−0.20 (-0.30, −0.10)	−14.61 (-14.97, −14.25)
Private motorized (passenger)	2.07 (1.73, 2.41)	−0.24 (-0.40, −0.08)	−14.16 (-14.75, −13.57)

MobiliSense Study, 283 participants, 7495 trip stages.
CI: confidence interval.
NA: Not Applicable.
Note: Minute ventilation was estimated from METs and was used to calculate the inhaled dose.
[a] The multilevel linear models included a random effect at the individual level. The crude and the detailed transport mode variables were introduced in separate models. Observations in the models were weighted for the duration of trip stages.
[+] All the models for breathing zone concentrations and inhaled doses were adjusted for seasons, weekdays vs. weekend, time of the day (morning, afternoon, evening, and night), and background concentration of $PM_{2.5}$ and NO_2 in GPS tracks based on Airparif monitoring stations.

with longer sitting times. Furthermore, waist worn accelerometer specifically underestimates METs for biking (Lopez et al., 2018), as it mostly detects the upper body motion which is limited while biking. This explains, unlike other methods, why MET-derived minute ventilation was lower for other active modes than for walking trip stages. It should be noted that the correlation coefficients and mean differences obtained in our study between all the ventilation estimation methods based on 1-minute level data (Appendix 6) are similar to what was reported by Dons et al in 2017 (Dons et al., 2017).

4.1. Strengths

The primary strength of this study is its innovative methodology involving an efficient algorithmic pre-processing of GPS data followed by a phone-based mobility survey along with a thorough manual correction and complementation of GPS tracks, in order to verify and validate the time and location of each trip stage start and end points. A detailed discussion of benefits and limitations of the aforementioned methodology is reported elsewhere (Chaix et al., 2019).

Our study used portable mobile sensors for measuring breathing zone concentrations of BC, since monitoring from fixed stations does not represent personal exposure during commuting, particularly during heavy traffic flow (Huang et al., 2012). The novelty of our study pertains to the combination of continuously collected BC sensor data with a GPS-based mobility survey providing timestamped information on activity contexts.

Until now, probably for technical and logistic difficulties in estimating ventilation rate in real-life settings from physiological measurements (heart rate, breathing rate, and acceleration), limited studies have reported inhaled doses of air pollutants in non-experimental settings especially across various transport modes. Among studies that assessed inhaled doses of BC across transport modes, none have adopted more than one method for the calculation of minute ventilation to

validate their findings (de Nazelle et al., 2012; Correia et al., 2020; Li et al., 2015; Tan et al., 2017; Morales Betancourt et al., 2019; Targino et al., 2018). Two studies among them did not calculate ventilation rate from their own participants' physiological characteristics (Correia et al., 2020; Targino et al., 2018). Therefore, our study significantly advances the knowledge of inhaled doses of air pollution during transport, by calculating inhaled doses using 4 different methods and comparing their discrepancies.

The present study represents an improvement in terms of generalizability compared to several previous work, by including non-experimental trips/routes made by the general public in their usual settings. Most of the previously conducted experimental studies included less than 500 trips (less than 200 for many of them) (de Nazelle et al., 2012; Correia et al., 2020; Li et al., 2015; Rivas et al., 2017; Jeong et al., 2017; Tan et al., 2017; Morales Betancourt et al., 2019; Targino et al., 2018; Velasco et al., 2019), while our study included more than 7000 trips from people observed in their usual life. Several of the previous studies only measured BC in specific hours such as peak hours, rush hours, or off hours, or only on week days (de Nazelle et al., 2012; Correia et al., 2020; Li et al., 2015; Rivas et al., 2017; Jeong et al., 2017; Tan et al., 2017; Morales Betancourt et al., 2019; Merritt et al., 2019; Targino et al., 2018), whereas all possible hours of days and days of weeks were considered in our study. Another generalizability issue in previous studies is that they were unable to include trips made in more than two seasons (de Nazelle et al., 2012; Correia et al., 2020; Li et al., 2015; Rivas et al., 2017; Jeong et al., 2017; Tan et al., 2017), unlike our study (which included 2487, 1904, 1488, and 1616 trip stages in Autumn, Spring, Summer, and Winter).

Our study distinguished between 8 different types of transport modes with a sufficient number of trips in each category, while previous studies considered at the most 4 transport modes with a lower number of trips per mode (de Nazelle et al., 2012; Correia et al., 2020; Rivas et al., 2017; Jeong et al., 2017; Tan et al., 2017; Morales Betancourt et al., 2019; Ham et al., 2017; Targino et al., 2018). Most of the studies did not consider walking as a separate mode of transport and did not examine the association of BC with this mode (Correia et al., 2020; Rivas et al., 2017; Apte et al., 2011). In 2019 Chaix et al. reported that 42.3% of the overall trips made over 7 days in the Paris metropolitan area was attributed to entirely walking (Chaix et al., 2019). Therefore; to improve the validity of our study and better inform national and regional policy makers about the real BC exposure while travelling; we included walking as a separate transport mode in our analysis.

Our study accounted for background variation of air pollutants, which is critical to compare exposure measurements made in different places and times. Our study integrating data from air quality models with continuous sensor measurements was useful to estimate the specific contribution of transport activity to personal exposure level as distinct from background concentrations.

4.2. Limitations

Inhaled dose in our study was calculated in two stages: 1) estimation of minute ventilation from physiological parameter (heart rate, breathing rate, and body accelerations), age, and sex of the participants and 2) calculation of inhaled dose as a product of estimated minute ventilation with air pollution concentration. These calculations involved several approximations which likely introduced intrinsic error in estimated doses.

Furthermore, as waist worn accelerometer is known to underestimate METs for biking (Lopez et al., 2018 Feb) and thus tends to assign underestimated minute ventilation to such travel mode (Dons et al., 2017), the inhaled dose of BC reported in our study for other active modes may underrepresent the real BC inhaled dose for these transport modes.

If specific trip stages in terms of BC exposure and/or minute ventilation were more frequently excluded for particular modes due to non-wear of any of the sensors, then this would bias the comparison of inhaled doses across the transport modes.

5. Conclusion

While higher breathing zone concentrations of BC were documented for all transport modes compared to walking and particularly in the metro and for drivers of private motorized vehicles, the highest inhaled doses were documented for other active modes, and to some extent for walking. Such exposure to BC in different transport modes is a threat for human health, e.g., for blood pressure (Mordukhovich et al., 2009; Liu et al., 2009) or in terms of reduction of lung function (Laeremans et al., 2018). While particles involved in the metro environment might include iron oxides (Moreno et al., 2015; Seaton et al., 2005), BC particles in the road environment are produced from the incomplete combustion of diesel fuel and might be highly toxic to human health (Kelly and Fussell, 2012).

This study adds evidence that choice of transport mode, ventilation rate, and duration of travel are significant predictors of breathing zone concentrations and inhaled doses of BC. Further studies are required to better elucidate the role of specific characteristics of transport microenvironments (e.g., ventilation systems used in private motorized vehicles, configuration of underground public transport environments), of the geographical location of the trips, and of the specific routes that are followed.

CRediT authorship contribution statement

Sanjeev Bista: Methodology, Software, Investigation, Formal analysis, Writing – original draft. **Clélie Dureau:** Project administration, Data curation, Resources. **Basile Chaix:** Funding acquisition, Conceptualization, Methodology, Validation, Writing – review & editing, Supervision.

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.

Acknowledgment

This work was supported by the European Research Council (ERC) under the European Union's Horizon 2020 research and innovation program (grant agreement 647000, 2014 ERC Consolidator grant, MobiliSense project). The funding institution had no role in the design of the study and collection, analysis, and interpretation of data and in writing the manuscript.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envint.2021.106990.

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