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Personal exposure to Black Carbon in transport microenvironments

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ABSTRACT

We evaluated personal exposure of 62 individuals to the air pollutant Black Carbon, using 13 portable aethalometers while keeping detailed records of their time-activity pattern and whereabouts. Concentrations encountered in transport are studied in depth and related to trip motives. The evaluation comprises more than 1500 trips with different transport modes. Measurements were spread over two seasons. Results show that 6% of the time is spent in transport, but it accounts for 21% of personal exposure to Black Carbon and approximately 30% of inhaled dose. Concentrations in transport were 2–5 times higher compared to concentrations encountered at home. Exposure was highest for car drivers, and car and bus passengers. Concentrations of Black Carbon were only half as much when traveling by bike or on foot; when incorporating breathing rates, dose was found to be twice as high for active modes. Lowest 'in transport' concentrations were measured in trains, but nevertheless these concentrations are double the concentrations measured at home. Two thirds of the trips are car trips, and those trips showed a large spread in concentrations. In-car concentrations are higher during peak hours compared to off-peak, and are elevated on weekdays compared to Saturdays and even more so on Sundays. These findings result in significantly higher exposure during car commute trips (motive 'Work'), and lower concentrations for trips with motive 'Social and leisure'. Because of the many factors influencing exposure in transport, travel time is not a good predictor of integrated personal exposure or inhaled dose.

1. Introduction

In dedicated studies, it has been shown that traffic exposure may trigger health effects like myocardial infarction (Brook et al., 2010; Mills et al., 2007; Nawrot et al., 2011; Peters et al., 2004). Black Carbon (BC), or other traffic-related pollutants correlated with BC (NO_2, CO, Elemental Carbon, Ultrafine particles), may also provoke short or longer term health effects, e.g. cardiovascular disease (Baja et al., 2010; Gan et al., 2011; McCracken et al., 2010), adverse respiratory health outcomes (Lin et al., 2011; McCreanor et al., 2007; Patel et al., 2010) or neurological effects (Bos et al., 2011; Power et al., 2011; Suglia et al., 2007). Recently it has been stressed by Janssen et al. (2011) that BC is a useful new indicator for the adverse health effect of traffic-related air pollution.

Typically epidemiological studies try to relate an exposure metric to certain health effects in exposed or less exposed people. If using a generic exposure metric like population exposure or air quality measured at one specific place, it neglects the large contrast and variation in personal exposures that is important in epidemiological studies. For example, individuals traveling from hot spot to hot spot or professional drivers will be exposed to far higher concentrations compared to a hypothetical static population. In previous studies using activity-based models or personal monitors it is demonstrated that the transport activity, although short in duration, can be responsible for quite a large part of integrated personal exposure to combustion-related pollutants (Beckx et al., 2009; Dons et al., 2011; Fruin et al., 2004; Marshall et al., 2006). Understanding the variation in exposure can contribute to a more accurate exposure assessment and reduce misclassification of air pollution health effects. This is of major importance when trying to define the health effects of pollutants that are highly variable in time and space, like e.g. traffic-related air pollutants (Setton et al., 2011; Strickland et al., 2011; Van Roosbroeck et al., 2008).

In the light of understanding the role of transport activities on total accumulated exposure, a large personal monitoring campaign was set up. BC was measured on a 5-min time resolution, allowing air quality data to be linked with reported activities. In this paper the focus will be on exposure in traffic microenvironments; however transport is always considered as part of a complete 24 h diary, enabling the identification of trip motives and the calculation of the contribution of transport to integrated exposure and inhaled

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dose. BC was chosen as a pollutant because of its relevance for health, and because of the availability of suitable measurement devices. Moreover the interest of policy makers in BC was aroused due to emerging evidence on health effects and the impact of BC on global warming. In developed countries, motorized transport, and mainly diesel vehicles, are considered to be the most important source of BC, whereas in developing countries biomass burning may be important (Highwood and Kinnersley, 2006; Kirchstetter et al., 2008).

2. Materials and methods

Personal exposure to BC is measured with portable aethalometers (microAeth Model AE51, (AethLabs, 2011)), carried by 62 individuals for 7 consecutive days. During the sampling, participants were urged to meticulously keep track of their executed activities by reporting them in an electronic diary fitted with a GPS. On top of that, a short questionnaire asked for characteristics of the individual, the household and the residence. More details on the configuration of the devices, quality assurance and data analysis can be found in Dons et al. (2011). Sixteen people took part in a pilot study in summer 2010; half of them participated again in a more elaborate campaign in winter 2010–2011. The other half was either unwilling or unable to participate a second time. The winter campaign was supplemented with 38 new volunteers. Because we wanted to focus primarily on the impact of the time-activity pattern on personal exposure, we measured two people sharing the same residence. In summer 2010 all 8 couples were measured sequentially; in the winter campaign a maximum of three couples were measured simultaneously each week, for eight weeks in a row.

Some small adaptations were made in the winter campaign compared to the summer. The PARROTS software installed on a small handheld computer (Kochan et al., 2010), to fill in executed activities and trips, was somewhat simplified to further reduce respondent burden, without significant data loss. In the first campaign couples consisted of a full-time worker and a homemaker or part-time worker; in winter we relaxed this constraint: there was no further limitation on the work schedule. Other adaptations all concerned quality assurance and quality control (e.g. additional comparison with filter-based EC analysis and with a Multi-Angle Absorption Photometer (MAAP) measuring BC simultaneously at the official air quality monitoring stations).

To maintain data integrity, we corrected the aethalometer readings in different ways. First, all data showing an error code were excluded from the dataset (except for low battery events). In addition, we excluded data when the attenuation was above 75, whereas the instrument only gives an error code if attenuation is around 100. The value of 75 is a conservative lower limit as proposed by Virkkula et al. (2007), Hansen (2005) proposed the range of 75–125 as a suitable advisory limit for aethalometers. Finally we did an inter-comparison between all 13 devices used, to correct for device specific deviations (see Supplemental material for details, corrections were between 1% and 23%). The sample flow of all instruments, set at 100 ml min^{-1}, was checked before the measurement campaign.

During the sampling campaign, data from a fixed BC monitor on a suburban background location (station 40AL01 – Antwerpen Linkeroever, operated by the Flemish Environment Agency) was used to correct for non-simultaneous measurements (for the methodology, see Supplemental material).

Negative measurements were included into the analysis, because a temporary false decrease in measured absorption is offset in the next observation(s) (McBean and Rovers, 1998; Wallace, 2005). Only deleting the negative values would overestimate average BC concentrations.

To calculate the contribution of each activity to dose, a translation of exposure to inhaled dose is made by defining a minute ventilation per activity and per transport mode; gender was also taken into account. Inhalation rates are based on Allan and Richardson (1998) and Int Panis et al. (2010) (Supplemental material Table S2).

SAS 9.2 was used for data processing and statistical analysis.

3. Results

3.1. Study characteristics and time-activity patterns

All 62 volunteers participating in the measurement campaign measured their personal exposure for 7 consecutive days, 24 h a day, on a 5-min time resolution. This resulted in 124,992 single measurements, or more than 10,000 h of data. Some technical failures or human errors resulted in a data loss of approximately 4%. After data cleaning (excluding measurements with high attenuation or error signal), 17% of all data was not considered for further analysis. This is a rather high number, but it was necessary to maintain data integrity and, because of the very large dataset, a conservative limit could be used setting a high standard for the data analysis.

All volunteers were nonsmokers and not exposed to second-hand smoke at home. Everyone was of working age and there was a small bias toward higher education. Most participants worked in an office, and everyone worked in an indoor environment. All 62 participants had a driving license, but not all couples owned a car. Participants were living in Flanders, Belgium (Supplementary Figure S4). An overview of personal and household characteristics is given in Table 1 and car attributes are summarized in Supplementary Table S3.

Based on the activity diaries, it was calculated that volunteers spend 6.3% of their time (90 min per day) in transport; 35.5% of the day is spent sleeping (Table 2). The majority of trips were by car; but one third of all travel time was by slow modes (bike, on foot). There are relatively more trips as car passenger in the weekend and on off-peak hours compared to car drivers. Train and metro are generally used by commuters, with a large share of trips in traffic peak hours and on weekdays. Trips as car driver, cyclist, bus passenger or walking are spread in the same way throughout the day and throughout the week. All light rail and metro trips are in urban areas, whereas car trips are often on highways (>25% of total time) and on rural roads (>30% of total time). More than 70% of the time, trips by bike or on foot are on urban or suburban roads.

Table 1
Characteristics of the study participants.

		Summer	Winter
Personal characteristics			
Gender[a]	Male	8	23
	Female	8	23
Year of birth[a]	1951–1960	2	6
	1961–1970	7	12
	1971–1980	5	14
	1981–1990	2	14
Education/Highest degree[a]	Primary or secondary school	2	3
	Higher education, non-university	6	10
	Higher education, university	8	33
Working status[a]	Full-time worker	8	32
	Part-time worker	3	8
	Non-worker	5	6
Household characteristics			
Average household size[a]		3.88	3.65
Average number of cars per household[a]		1.38	1.48

[a] Results based on questionnaires filled in by the participants.

Table 2
Time-activity pattern, contribution of each activity to total BC exposure, and contribution to inhaled dose (average of 62 participants).

Activity type	Time-activity pattern	Contribution to exposure	Contribution to dose
Home-based activities	29.9%	26.7%	21.8%
Sleep	35.5%	25.0%	13.9%
Work	17.0%	12.2%	12.8%
Social and leisure	6.3%	8.9%	12.8%
Shopping	1.1%	2.0%	3.2%
Other	3.9%	4.3%	5.6%
In transport	6.3% (100%)	21.0% (100%)	29.8% (100%)
Car driver	2.9% (45.3%)	12.3% (58.6%)	10.5% (35.2%)
Car passenger	0.7% (11.4%)	2.2% (10.3%)	1.7% (5.8%)
Bike	1.0% (15.7%)	2.5% (11.8%)	9.1% (30.5%)
On foot	1.0% (16.4%)	2.2% (10.5%)	6.6% (22.3%)
Train	0.5% (7.9%)	0.9% (4.2%)	0.9% (3.0%)
Light rail/metro	0.1% (0.8%)	0.2% (1.0%)	0.2% (0.7%)
Bus	0.2% (2.4%)	0.7% (3.6%)	0.7% (2.5%)

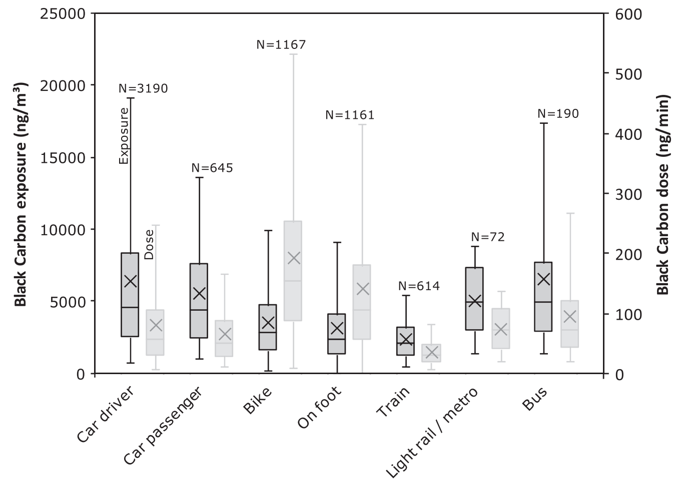

Fig. 1. Personal exposure (black box plots) and dose (gray box plots) in different transport modes. The 'N = ' indicates the number of 5-min observations used to calculate the average and the percentiles. Represented are P5, 1st quartile, median, 3rd quartile and P95. The cross marks the average.

3.2. Personal exposure measurements

Average personal exposure was 1592 ng m^{-3}, with a standard deviation of 468 ng m^{-3}. This is comparable with the average concentration measured by the fixed suburban monitor (1620 ng m^{-3}). The volunteer with the lowest personal exposure was exposed to 652 ng m^{-3} on average and the highest exposed participant to the study had a personal exposure of 2773 ng m^{-3}. BC concentrations are lognormally distributed within each participant, meaning that there are a lot of 5-min observations with relatively low concentrations, and some observations where participants are highly exposed.

Lowest average concentrations were observed during home-based activities (1360 ng m^{-3}), working (1077 ng m^{-3}) and sleeping (1090 ng m^{-3}). The highest average concentrations by far, were encountered while in transport (5132 ng m^{-3}). High peaks are especially prominent in transport: 95th percentile is 15,569 ng m^{-3}. Elevated average concentrations of 2445 ng m^{-3} and 2540 ng m^{-3} are observed for social and leisure activities, and for shopping respectively. Looking at microenvironments instead of activities, shows the same trends: high exposure in transport, lowest concentrations in private homes (1255 ng m^{-3}) and at work locations (1068 ng m^{-3}). Concentrations in transport were 2–5 times higher compared to concentrations encountered at home (Figure S5 and Figure S6). Although the amount of time spent in transport is relatively small, this nevertheless corresponds to 21% of personal exposure to BC due merely to the high concentrations measured in transport (Table 2).

The transport category can be subdivided in different classes according to transport mode (Fig. 1). Lowest concentrations were measured in trains, with a mean of 2394 ng m^{-3}. Volunteers traveling with slow modes, by bike or on foot, were confronted with higher average exposures ranging from 3175 ng m^{-3} to 3555 ng m^{-3}. It should be noted that the average exposure of cyclists and pedestrians was 62% lower when the trip was a leisure trip, indicating the use of alternative routes instead of using the shortest (not seldom the most polluted) route as is more often the case for commute trips (Dons et al., 2011). The exposure of volunteers traveling by motorized transport was highest (car driver: 6432 ng m^{-3}; car passenger: 5583 ng m^{-3}; bus passenger: 6575 ng m^{-3}; and light rail/metro passenger: 5066 ng m^{-3}). The results of the light rail/metro category are indicative since they only encompass 23 trips, although spread over different cities and weeks. In summary, the exposure-ratios of BC in different transport modes, in typical Belgian conditions, are: automobile:bicycle ratio = 1.77; automobile:foot ratio = 2; automobile:bus ratio = 0.96; and automobile:train ratio = 2.63.

More than 65% of all trips were made either as car driver or as car passenger. An important determinant of the concentrations measured inside a car is the timing of a trip. Fig. 2 shows the hourly variation in average concentrations: highest in-vehicle concentrations are observed during traffic peak hours (morning peak between 7 and 10 a.m., evening peak between 4 and 7 p.m.). This trend is less pronounced in other transport modes, probably because traffic congestion on rush hours affects car users more than e.g. cyclists or pedestrians. The impact of time-of-day on concentrations in microenvironments different from transport, is more limited although concentrations are still 33% higher in the evening (17–22 h) than during the day (8–16 h).

Day of the week affects personal exposure encountered in a car. We corrected the personal measurements for daily differences in background concentrations, but still differences between days appeared (Fig. 3). Between working days, there are only minor differences in in-vehicle concentrations (from 5366 ng m^{-3} on Wednesday, to 5893 ng m^{-3} on Thursday), but they are in line with the traffic intensity on these days. Concentrations are lower on Saturdays (4459 ng m^{-3}) and Sundays (3830 ng m^{-3}). This difference between in-car concentrations on weekdays and weekend days was found to be statistically significant. A paired test was used to limit the influence of specific characteristics of each car: we assume that individuals drive the same car on all days of the week.

3.3. Inhaled dose

Inhaled dose of the participant with the lowest average exposure to BC was 14,134 ng day^{-1}. The highest exposed individual inhaled on average 77,698 ng day^{-1}. In our study, the lowest and highest exposed individual also had the lowest and highest BC intake (dose), but this is not necessarily the case and depends on the executed activities.

When comparing exposure and inhaled dose in Fig. 1, it immediately shows up that the active modes contribute more to inhaled dose than to exposure. The highest dose is encountered on a bike, with an average dose of almost 200 ng min^{-1}. For inhaled dose, the ratios between different transport modes become: automobile:bicycle ratio = 0.41; automobile:foot ratio = 0.56; automobile:bus ratio = 0.82; and automobile:train ratio = 2.16.

On average the relative importance of the transport activity increases, up to 30%, when incorporating inhalation rates (Table 2).

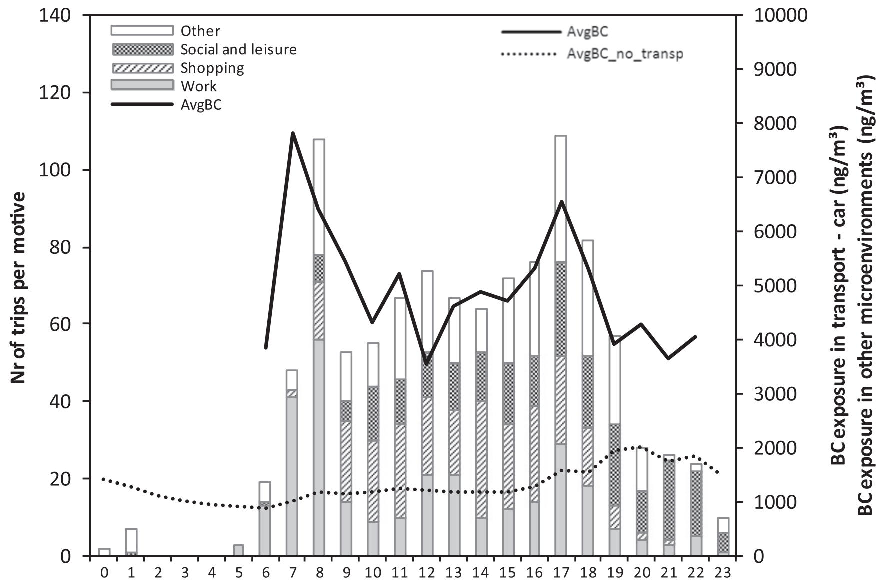

Fig. 2. Timing of a car trip and the impact on in-car BC concentrations. The black line shows the in-vehicle concentrations for every hour of the day. Each trip is assigned to the start hour of the trip. If less than 10 trips are available, the average was not included. The dashed line represents the average BC concentrations in all other microenvironments, excluding transport. The bars indicate the number of car trips and the corresponding trip motives on which the average concentration is based.

This difference is due to lower respiration during sleeping and higher respiration during active travel (e.g. on a bike minute ventilation is 6–7 times higher compared to ventilation during sleep). The average daily dose incurred during transport is therefore similar to the average dose incurred from home-based activities, and much larger than the inhaled dose during the sleep activity.

3.4. Approximations for exposure and dose

Because multiple factors influence exposure in transport, it is not straightforward to relate a simple metric such as travel time to integrated personal exposure or inhaled dose (Fig. 4). We demonstrated that average exposure in transport is very high, but also

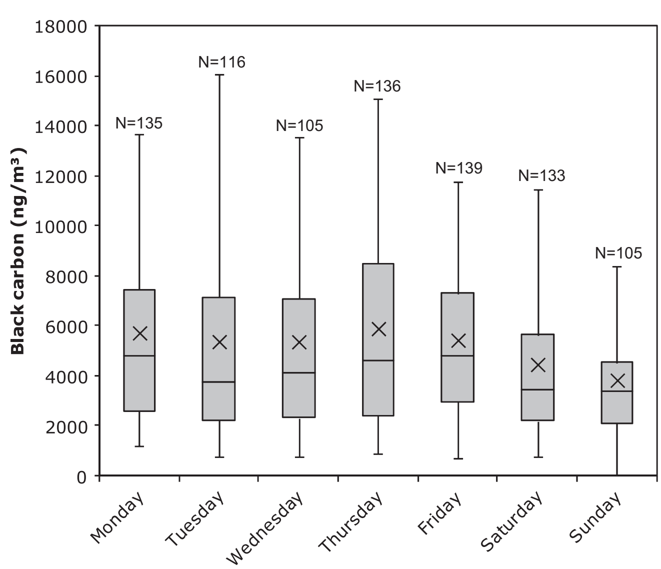

Fig. 3. Timing of a car trip (day of the week) and the impact on in-car BC concentrations. The '$N =$ ' indicates the number of trips used to calculate the average and the percentiles. Represented are P5, 1st quartile, median, 3rd quartile and P95. The cross marks the mean value. In-car concentrations are significantly lower in the weekend than on weekdays (Paired T-test $p < 0.01$).

highly variable between individuals depending on the transport modes used, the timing of trips (time-of-day, day of the week), and possibly the geographical location of the trip (further research). Limiting travel time to travel time by car reveals an equally poor correlation with personal exposure (Figure S7). On the other hand, our results show a better correlation between average exposure in transport and average personal exposure (Figure S8). Because we know that trips are responsible for 30% of daily inhaled dose, the relationship between travel time and dose is expected to be somewhat better: with 6% of explained variance, travel time is not very predictive for inhaled dose either. For comparison, the correlation between residential outdoor concentrations and personal exposure is 0.32 (Figure S9).

4. Discussion

In our previous paper (Dons et al., 2011) we already indicated that transport is responsible for almost a quarter of accumulated exposure, although individuals travel no more than 6% of the day. The results of the present more elaborate study confirm that transport indeed accounts for 21% of personal exposure. Our results further show that transport contributes up to 30% to the inhaled dose. In spite of the limited time spent in transport, more BC particles are inhaled during, on average, 90 min of transport compared to e.g. >500 min of sleeping or even during all other home-based activities combined. Nevertheless many epidemiological and HIA studies only include residential exposure and ignore exposure in transport; just recently several studies started to assess the health effects of in traffic exposure to air pollution (McCreanor et al., 2007; Strak et al., 2010; Zuurbier et al., 2011).

The levels of exposure in transport depend on several factors; some of which were studied in depth in this paper: transport mode choice and timing of a trip. The most obvious factor influencing exposure during travel is transport mode choice. The highest average concentrations are encountered in motorized transport: car and bus. This is in line with findings of Adams et al. (2002) measuring Elemental Carbon in different transport modes, and with the review paper of Kaur et al. (2007) stressing the general trend in multi-mode exposure assessments on fine particulate

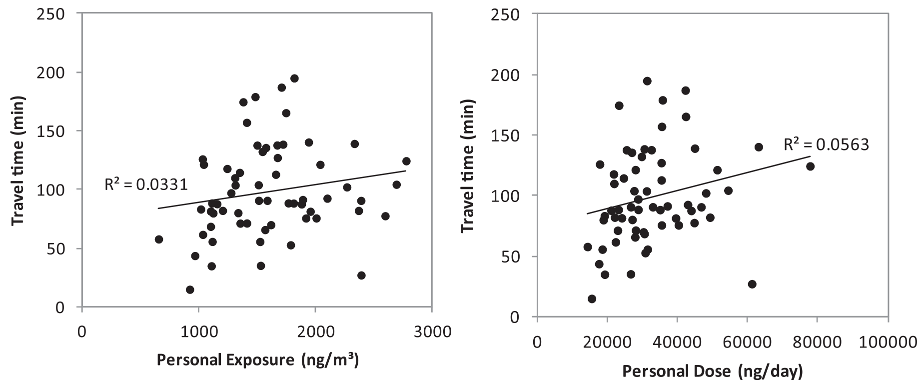

Fig. 4. Correlation between personal exposure, daily dose and travel time. Each mark represents one of 62 volunteers.

matter: pedestrians and cyclists experience lower exposure concentrations than individuals inside vehicles. Zuurbier et al. (2010) measured soot in three different transport modes: car, bus and bicycle. Soot levels were highest in cars and buses, and lowest by bike along a low-traffic street. A review on ultrafine particle (UFP) exposure in the transport microenvironment, encompassing approximately 3000 individual trips in total, but originating from different studies, found overall mean UFP concentrations of 3.4; 4.2; 4.5; 4.7; 4.9 and 5.7 $\times$ 10^4 particles cm^{-3} for the bicycle, bus, car, rail, walking and ferry modes, respectively (Knibbs et al., 2011). Boogaard et al. (2009) and Int Panis et al. (2010) reported smaller and inconsistent car/bicycle ratios for UFP in different towns. Particle number counts (PNC) were 5% higher on average in cars than on bicycles but this hides important local differences in either direction. In-vehicle BC concentrations were measured during 29 car trips by Rodes et al. (1998). Concentrations ranged from below the detection limit to 23,000 ng m^{-3}; with an average of 6000 ng m^{-3} in urban Los Angeles, and 4000 ng m^{-3} as the statewide in-car average (Fruin et al., 2004). Sabin et al. (2005a; 2005b) measured BC levels inside school buses between 900 ng m^{-3} and 19,000 ng m^{-3}; this is comparable to concentrations measured in this study although our measurements were not limited to school buses or school bus hours. Bizjak and Tursic (1998) measured BC in buses as well and found higher concentrations in diesel buses (10,000–50,000 ng m^{-3}), and lower concentrations inside a new gas-powered bus (5000–15,000 ng m^{-3}). The portable aethalometer model AE51 was used by Weichenthal et al. (2011) to measure concentrations on a bike. Two types of routes were cycled: a high-traffic route (mean = 2520 ng m^{-3} (range 890–5670 ng m^{-3})) and a low-traffic route (mean = 1079 ng m^{-3} (range 173–3197 ng m^{-3})). These concentrations are lower than our mean concentrations for cyclists, but the trips in the study of Weichenthal et al. (2011) all took place outside peak hours (between approximately 11.30 a.m. and 12.30 p.m.), only in the warmer season and in an area with lower background concentrations. Apte et al. (2011) used the aethalometer model AE51 to determine BC exposure in auto-rickshaws in New Delhi, India. The geometric mean was 42,000 ng m^{-3}, which is several times higher than BC concentrations observed in high-income countries. No studies were identified measuring exposure to BC in multiple modes, although the last study mentioned some limited in-car measurements. Because in Fig. 1 the results were corrected for changing background concentrations, a direct comparison of absolute concentrations might be difficult (uncorrected concentrations are presented in Table S1). The overall ratios between modes of transport, presented in the previous section, can be biased by the fact that some modes of transport might be used preferentially in conditions that also affect ambient BC concentrations, e.g. in specific locations, rural versus urban, workdays versus weekend days, time-of-day or weather conditions. In Table S4 and S5 exposure-ratios are split up for morning rush hours, evening rush hours and non-rush hours. The ratio automobile:(slow modes) is lower on non-rush hours which probably reflects the fact that traffic congestion affects car users more than cyclists and pedestrians, as they are directly exposed to exhaust from preceding cars.

Clear differences in exposure between different transport modes appeared in our measurement campaign, but the large variation in BC concentrations in each transport mode reveals that multiple factors affect these concentrations. The timing of a trip seemed an evident, but little explored, parameter influencing in-transit concentrations. We saw elevated in-car concentrations on traffic peak hours, compared to off-peak hours; and elevated levels on working days compared to the weekend. Apte et al. (2011) looked at day of week differences in BC exposure; in line with our results, they found no consistent differences between working days. Sampling was limited to Monday through Friday, so any weekend effects could not have been detected. They did find time-of-day trends for BC: concentrations were 32% lower during the morning commute as compared to the evening commute, mainly attributable to the on-road environment. Hertel et al. (2008) looked at the impact of time-of-day on NO_x concentrations on bike and bus. Higher than average concentrations were observed during morning rush hour compared to off-peak hours. The evening rush hour peak was less pronounced, probably because the period with high traffic intensities was also spread over more hours. The time-of-day variation in concentrations was mainly observed in buses on highly trafficked streets; the trend was less noticeable for bike trips on quieter streets. From Fruin et al. (2004) we know in-vehicle BC concentrations are highest when directly following diesel-powered vehicles. Considering the large number of diesel vehicles on Belgian roads (over 60% of all private cars in Belgium are diesel (NIS, 2010), a number that is remarkably higher than in neighboring countries), there is a high chance that participants drove behind diesel vehicles during rush hour. According to Westerdahl et al. (2005), on-road BC concentrations also appeared to increase sharply as diesel truck traffic increased. A high proportion of traffic in Belgium is transit diesel truck traffic, especially on highways.

Trip motive can be defined as the activity that is performed on the destination side of a trip unless this is a 'home-based activity': in that case the activity at the origin side is defined as the trip motive (Cools et al., 2011). From Fig. 2 it became clear that there are many work trips in the morning peak hours and there is a large share of leisure trips in the evening. Fewer trips have motive 'work' in the evening peak hour because of trip chaining. If we combine previous findings, it is straightforward to conclude that average in-car exposure is highest for trips with motive 'Work' (Figure S10):

these trips are often in traffic peak hours and on weekdays; and we know that during these time-periods the in-car concentrations are highest. Leisure trips are mainly in the evening or in weekends, and thus lower average exposure can be expected.

It is dose that is directly linked to health endpoints and thus relevant in epidemiological studies. Nevertheless many of these studies derive exposure and exposure-response functions based on the residential location only. In our study, we found that participation in traffic, especially active transport as a cyclist or pedestrian, increases inhaled dose more than proportionally. On average inhaled dose is clearly larger for slow modes compared to all other modes, but on an individual basis the height of the dose depends on several factors. As already stated, it is very important to take timing and location into account. Breathing rates can also differ by individual and by the degree of physical exertion. Assumptions on minute volume were made based on literature and are an approximation of real minute volume. The question remains whether short periods exposed to elevated concentrations have any significant health effect on an individual. Weather (rain or heavy wind) might be a factor that shifts cyclists to days with higher BC concentrations. On the other hand, cyclists themselves try to avoid busy traffic or hilly terrain, take the least polluted and thus 'healthiest' route, and that way they decrease their exposure to traffic-related air pollution (Hertel et al., 2008; Int Panis et al., 2010; Zuurbier et al., 2010). Several studies already demonstrated that potential health effects caused by air pollution in traffic, are more than offset by the positive effects of active travel (de Hartog et al., 2010; Rabl and de Nazelle, 2012; Woodcock et al., 2009).

Integrated weeklong personal exposure is highly variable between individuals, ranging from 652 ng m^{-3} to 2773 ng m^{-3}. Those individual differences are much larger than population based estimates that take activities into account (Beckx et al., 2009). Accurate exposure assessment, focusing on personal exposure rather than population based estimates, is thus critical to further reduce exposure misclassification in epidemiological studies. Personal measurements, as shown in this paper, are one way of estimating personal exposure more accurately. However, a methodology taking background variation into account is essential to compare measurements performed at different times. A combination of air quality models and activity-based models, producing time-activity schedules for every individual actor in a population, seem promising since models will be less hampered by sample size limitations than measurements. Using an activity-based model, exposures can be turned into inhaled doses in a straightforward way by assigning a minute ventilation to every activity.

It was impossible to relate in-vehicle concentrations to certain characteristics of the car since it is not known which trip is undertaken by exactly which car. This is a major drawback of this study, together with the lack of control over the routes taken, and the ventilation settings of the car. On the other hand we randomly selected different cars that were driven in a real-life setting, and from the questionnaires we had some basic information on the cars. This resulted in a very large and representative dataset, e.g. trips of different durations, trip chaining behavior was included, trips were geographically dispersed over a wider region, etc. The choice of volunteers participating in this study was somewhat biased, although a comparison of the activity diaries with a large cohort (over 1600 families) of the Flemish Travel Behavior Survey reveals good correspondence (Cools et al., 2011). Time in transport and the percentage of time in each transport mode, are also comparable to the Flemish average. We used only one monitor to correct for changing background concentrations; these measurements may not be completely representative for background concentrations in the entire study area. Unfortunately other fixed monitors, e.g. on rural background locations, were not available at the time of the measurements. Seasonal effects on BC exposure were not considered because we scaled the background-part of all observations based on concentrations measured at a fixed suburban background monitor. In contrast, we did observe a weekend – weekday effect because it was unrelated to changes in background concentrations.

In our study highest exposures were observed during traveling (especially car driving). Transport mode choice and timing of a trip proved important variables influencing exposure in transport. It was demonstrated that trips with motive 'work' had highest average in-car concentrations because they are mainly driven during peak hours. Inhaled dose per minute is highest during cycling and walking, but can be influenced by taking an appropriate route. We provided evidence that travel time is an unsatisfactory parameter to predict personal exposure to BC.

High peak hour concentrations may, at least partially, be caused by more frequent use of highways or urban roads (Sabin et al., 2005a). Westerdahl et al. (2005) suggested an impact of diesel truck volume on BC concentrations inside vehicles and the speed of a car may have an impact on the air exchange rates (Fruin et al., 2004), thereby influencing in-vehicle exposure to particles. Further research will therefore focus on these aspects as well as on the geographical location and road characteristics of the trips.

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Appendix. Supplementary material

Supplementary data related to this article can be found online at doi:10.1016/j.atmosenv.2012.03.020.

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