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Transport most likely to cause air pollution peak exposures in everyday life: Evidence from over 2000 days of personal monitoring

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ABSTRACT

Background: Air quality standards are typically based on long term averages – whereas a person may encounter exposure peaks throughout the day. Exposure peaks may contribute meaningfully to health impacts beyond their contribution to long term averages, and therefore should be considered alongside longer-term exposures. We aim to define and explain peak exposure to black carbon air pollution and look at the relationship between short peak exposures and longer term personal exposure.
Methods: A peak detection algorithm was applied to pooled data from two independent studies. High-resolution personal black carbon monitoring was performed in 175 healthy adult volunteers for a minimum of two 24-h periods per person. At the same time, we retrieved information on the time-activity pattern. Data covered Belgium, Spain, and the United Kingdom. In total, 2053 monitoring days were included.
Results: Exposure profiles revealed 2.8 ± 1.6 (avg ± SD) peaks per person per day. The average black carbon concentration during a peak was 4206 ng/m^3. On 5.5% of the time participants were exposed to peak concentrations, but this contributed to 21.0% of their total exposure. The short time in transport (8%), was responsible for 32.7% of the peaks. 24.1% of the measurements in transport were categorized as peak exposure; while sleeping this was only 0.9%. When considering transport modes, participants were most likely to encounter peaks while cycling (34.0%). Most peaks were encountered at rush hour, from Monday through Friday, and in the cold season. Gender and age had no impact on the presence of peaks. Daily average black carbon exposure showed only a moderate correlation with peak frequency (r = 0.44). This correlation coefficient increased when considering longer term exposure to $r > 0.60$ from 10 days onward.
Conclusions: The occurrence of peaks varied substantially over time, across microenvironments and transport modes. Daily average exposure was moderately correlated with peak frequency. Real-time air pollution alerting systems may use the peak detection algorithm to support citizens in self-management of air pollution health effects.

1. Introduction

Traffic emissions are an important source of air pollution in cities. Short-term and long-term exposure to traffic-related air pollution have been associated repeatedly to a number of health outcomes in epidemiological studies (HEI, 2010). There is also toxicological evidence that acute exposure events, or peaks, have different and independent health effects compared to longer-term exposure averages (Smith, 2001; Zhou et al., 2017). Peak exposures may provoke immediate physiological changes, trigger a next stage in the development

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Table 1
Summary of the datasets used for the detection of peak exposures. Data were pooled before analysis.

Dataset	Reference	Study description	Person-days with at least 90% complete data (N = 2053)	Unique participants (N = 175)	Sex: Male (81 (46%))	Age (years) (36.2 (9.8))	Black carbon, 24-h personal exposure (1364 (738) ng/m^3))
Couples in Belgium	Dons et al. (2012)	24 h personal monitoring, 7 days continuously, 8 participants participated in 2 seasons, other participants only in 1 season; participants living in the region Flanders (Belgium); AE-51 (Aethlabs, USA) for black carbon, electronic diary for time-activity pattern; 2010–2011	344	54	27 (50%)	38.8 (10.1)	1474 (686) ng/m^3
Healthy adults in Europe (PASTA project)	Dons et al. (2017); Laeremans et al. (2018)	24 h personal monitoring, 7 days continuously, repeated in 3 seasons; participants living in Antwerp (Belgium), Barcelona (Spain), London (UK); AE-51 (Aethlabs, USA) for black carbon, SenseWear (BodyMedia, USA) armband for physical activity and time-activity pattern; 2015–2016	1709	121	54 (45%)	35.6 (9.6)	1342 (746) ng/m^3

Data are mean (SD) or N (%).

of a disease or even trigger myocardial infarction or death (Chen et al., 2017; Knibbs and Morawska, 2012; Lanki et al., 2006; Madsen et al., 2012; Nawrot et al., 2011; Peters et al., 2004; Rappaport, 1991). In addition, acute effects can cause discomfort (e.g. wheezing or dyspnea) resulting in temporary disability (Tian et al., 2017; Wegman et al., 1992). Repeated peak exposures may also contribute disproportionally to longer-term outcomes, due to either non-linear exposure response functions, or because repeated acute impacts overload protective or repair mechanisms (Knibbs and Morawska, 2012; Michaels and Kleinman, 2000; Zhou et al., 2017). As a consequence, people receiving a similar dose of pollution (either with or without peaks) could experience a different impact on their body.

Until now, epidemiological and especially occupational studies have assumed that (1) longer term averages is what matters most for chronic impacts, and (2) longer term averages and (repeated) peak exposures are sufficiently correlated. This would mean that a person experiencing many spikes in their exposure profile, would also be exposed to elevated average exposures over the same time interval. It is an open question whether this association holds in the context of 24-h continuous exposure while performing routine activities, with changing background concentrations and quickly changing exposures when switching microenvironments.

Peak exposure is a widely-used term to indicate increased exposure, and although intuitively clear, there is no fixed definition (Smith, 2001). A peak or spike could be defined as at least one time window of predefined length with an average exposure above a predefined threshold. Nieuwenhuijsen et al. (1996) defined a peak as a relatively short term period of which its exposure level is considerably higher than the exposure level of a long term period within which it occurs. Different parameters can be defined when looking at peak exposures, of which magnitude, frequency, and rate of increase seem to be the most relevant when considering health outcomes. A threshold can be operationally defined as concentrations higher than the 90th or 95th percentile over a fixed period (Jeong and Park, 2018; Maciejewska et al., 2015; Peters et al., 2014), or concentrations that deviate at least 50% or x standard deviations from an otherwise steady state concentration (Michaels and Kleinman, 2000). Alternatively, a threshold can be defined as a concentration below which no detectable physiologic response occurs (Preller et al., 2004; Wegman et al., 1992). Peaks have variable lengths with no agreed pre-defined durations; and the ability to detect a peak is often limited by the low time resolution of air pollution measurements. Frequency can be expressed as the absolute number of peaks per sample, but because sampling durations may vary, the number of peaks per hour or per day can be used instead as a normalized index (Preller et al., 2004). Finally, the rate of increase needs to be sufficiently rapid (i.e. sharp increase of exposure over a short timespan) to evoke a physiologic response: acute effects are likely to be dependent upon the rate at which the agent accumulates in the target organ (Wegman et al., 1992).

Mobile air pollution sensors enable us to measure personal exposure with a high temporal resolution, and to assess both peak and average exposure. Black carbon (BC; a component of fine particulate matter) is a pollutant that is often used as a good measure of traffic-related air pollution, and a portable device is available to measure time-integrated BC concentrations. According to the World Health Organization (Janssen et al., 2012), epidemiological studies provide sufficient evidence of an association of daily variations in BC concentrations with short-term changes in health, and there is evidence of associations of all-cause and cardiopulmonary mortality with long-term average BC exposure. In a large dataset of over 2000 days of personal monitoring of BC in three European countries, we aim to define and describe average and peak exposures; we study the role of personal characteristics, including time-activity and travel patterns, in explaining exposures to peaks; and we associate average exposure to the frequency of peaks.

2. Materials and methods

2.1. Study design

To get a sufficiently large and diverse sample, we combined data from two studies with a similar study design (Table 1). In brief, the studies performed 24-h personal monitoring of BC air pollution at a high temporal resolution, and also included some measure of the time-activity pattern either through a time-activity diary or through an activity tracker. The measurements were performed between 2010 and 2016. The first study was in a convenience sample of 27 couples living in Belgium (Dons et al., 2012); the second study included a total of 121 healthy adults living in Antwerp (Belgium), Barcelona (Spain), or London (UK), opportunistically recruited through the PASTA project (Dons et al., 2017; Gaupp-Berghausen et al., 2019; Laeremans et al., 2018). Professional drivers and people with high occupational exposures to BC were excluded from participation at recruitment. The pooled dataset consisted of 175 unique participants, and 2053 days with at least 90% of data available. Participants were mostly Caucasian and highly-educated.

Both studies measured personal exposure to BC using the microAeth type AE51 (AethLabs, San Francisco, CA, USA). This real-time pocket size instrument analyses BC by measuring the rate of change in absorption at 880 nm of transmitted light on a filter strip. The time resolution was set at 5 min; this is an integrated measurement that estimates the accumulation of black carbon particles during the previous 5 min. The flow was set at 100 ml/min. BC measurement data was treated similarly in both studies by starting from the raw data files. The Optimized Noise-reduction Averaging (ONA) algorithm was applied to reduce noise in the data and remove erroneously high or low values (Hagler et al., 2011). This smoothing process caused some 5-min observations to be averaged over longer time periods: in the first dataset 67% of the 5-min measurements were kept, 19% was averaged over 10 min, and 5% over 15 min; in the second dataset this was 59%, 20% and 8% respectively. A small minority of observations were averaged over longer time periods. Additionally, measurements that generated a filter saturation or flow error were removed, as well as BC values below $-50,000$ ng/m^3 or above 250,000 ng/m^3 that sporadically remained after the ONA algorithm was applied (0.01% of the cases). To assure data integrity, parallel measurements with all devices were performed prior and post to each study, including also a prior flow calibration (Dons et al., 2012).

The first study used an electronic diary for participants to report their activities and trips on a 5-min time resolution. The other study used the SenseWear (BodyMedia, USA) armband that automatically classified activities based on the internal accelerometer and propriety algorithms. An aggregated classification was decided on for the analysis including three activity types (sleeping, daily activities, transport) and three transport modes (walking, cycling, car or public transport).

2.2. Peak detection algorithm

BC peaks were signalled in the pooled dataset using a peak detection algorithm (van Brakel, 2014). The algorithm was initially developed for detecting sudden peaks in real-time timeseries data. The algorithm is suited for data with basic noise around a stationary mean, and with occasional peaks that deviate significantly from the noise, however, the width or height of the peaks is not pre-defined.

The algorithm starts by calculating a moving mean for BC beginning at midnight (Fig. 1). If the next observation is x standard deviations away from the moving mean, the algorithm sets this observation as a peak. A peak can be given a weight below one in the moving window to limit the impact of peak events on the mean.

By changing the parameters (time window, threshold, weight), the algorithm can be made more or less sensitive to peaks. A 2-h time window was chosen to set a stable baseline during night-time hours: we assumed this value to be representative of background air pollution, and a somewhat longer time window should limit the impact of short exposure peaks during the day. We favored a higher threshold, in this case 3.5 times the standard deviation, as we believe the rate of increase needed to be sufficiently rapid to be biologically relevant. Finally, a weight of 0.1 was applied as we acknowledge that the stationary mean may change when changing microenvironments – we're not interested in these changes in background concentrations, but rather in deviations from the new background. We additionally required peaks to have a minimum duration of 10 min (at least two separate observations with the 5-min time resolution of the BC air pollution monitor), and a local BC maximum within a peak of at least 2500 ng/m^3. The minimum duration of 10 min was introduced, firstly, to minimize the chance of detecting false peaks since a single reading may be impacted by mechanical shocks, or by rapid changes in humidity or temperature, and secondly, we hypothesize that the biological relevance increases. The value of 2500 ng/m^3 is an equivalent of 10% of the 24-h air quality guideline of the World Health Organization for PM2.5, knowing that about 10% of the PM2.5 fraction was identified as BC in urban environments in Europe (Maciejewska et al., 2015; Putaud et al., 2010; WHO, 2005). Sensitivity analyses were performed on the parameters (see below).

Negative peaks, i.e. observations below -3.5 standard deviations, were not frequent (0.8 ± 1.0 negative peaks per day) and were not treated as peaks further on in the analyses. Each 24-h period was analyzed separately and is referred to as a 'person-day'.

2.3. Data analysis

We first described the combined dataset using descriptive statistics. Magnitude, duration and frequency of BC peaks, and the contribution of peaks to integrated daily exposure were calculated. The 5-min measurements were summarized by several characteristics: activity type, transport mode, hour of the day, weekday, season, gender, age, and study area and dataset. We explored how those characteristics impacted peaks in the exposure profile. All potential variables that explained peaks were also entered in a mixed effects logistic regression model. We used a binary outcome variable: an observation was either a BC exposure peak or it was not. The person, and the dataset the observation belonged to, were included as random effects. All models were adjusted for activity type (sleeping/daily activities/transport), transport mode (walking/cycling/car or public transport), rush hour (yes/no), weekend (yes/no), season (spring/summer/fall/winter), and current age (continuous).

Average and cumulative exposure was related to frequency of peaks and cumulative peak exposure (duration * BC concentration during the peaks) over time intervals of 24 h with Pearson's r. To check whether the correlation coefficient increases when including multiple measurement days per person, BC exposures and number of peaks were averaged over multiple days per person, ranging from two days and up to 18 days, which is the maximum number of measurement days per person in our pooled sample. Additionally, we grouped person-days in four groups according to average BC exposure and number of peaks. Group 1 was defined as having a higher than average BC exposure, and a higher than average number of peaks (high exposure-many peaks). Group 2 had a lower than average BC exposure, but a higher than average number of peaks (low exposure-many peaks). Group 3 had a low average BC exposure, and a low number of peaks (low exposure-few peaks); and group 4 was defined as having a higher than average BC exposure, and a low number of peaks (high exposure-few peaks). We investigated whether a person was likely to be in the same group on multiple days (Chi-squared test), and whether peaks were triggered by other factors in the different groups.

As a test of sensitivity, it was checked whether using a simple, fixed threshold of 2500 ng/m^3 (also with a minimum peak duration of 10 min) or changing the parameter values in the peak detection

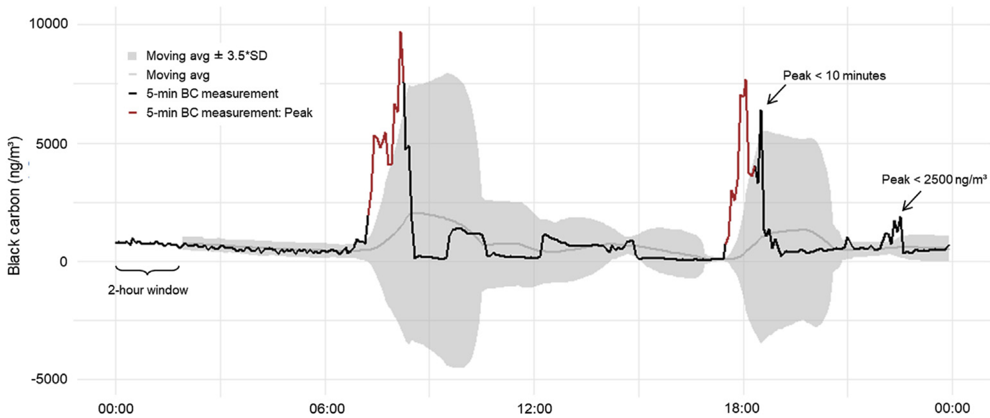

Fig. 1. Illustration of the peak detection algorithm in one 24-h timeseries (time window = 2 h; threshold = 3.5*SD; weight = 0.1). The timeseries presents personal monitoring of BC on May 21, 2015 in Barcelona, Spain. In this 24-h timeseries the algorithm detected two peaks (in red in the graph). The average BC concentration was 944 ± 1437 ng/m^3. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

algorithm would change our findings. More robust measures of scale were tested as well: the moving average was replaced by the moving median, and the standard deviation was replaced by the median absolute deviation (MAD).

Peak detection and all statistical analyses were performed in R statistical software.

3. Results

3.1. Description of the pooled sample

The pooled sample consisted of 2053 person-days from 175 unique participants between 18 and 62 years old. Each participant provided measurements during an average of 11.7 ± 4.7 days. This number was higher in the PASTA sample (14.1 ± 3.0), and lower in the couples (6.4 ± 2.8). The number of days per person ranged from 2 to 18.

The 5-min BC measurements (N = 578,885) were lognormally distributed (median 854 [IQR 464, 1513] ng/m^3) (see supplemental information). This already indicated the presence of peak exposures. About 30% of all measurements were made while participants were sleeping, and the median BC concentration was lowest during this activity (Table 2). 8% of all observations were categorized as transport, of which 42% walking, 40% in cars or on public transport, and 18% bicycling. The remaining fraction of time was spent doing daily activities (work, home duties, leisure, etc.). The 95th percentile concentration per activity was highest while in transport (8942 ng/m^3) and lowest while sleeping (2574 ng/m^3). The value of 2500 ng/m^3 which we have chosen as the minimum height for peaks corresponds to the 89th percentile.

3.2. Peak exposure

An average number of 2.8 ± 1.6 peaks per day were detected by the peak detection algorithm, with a maximum of 10 peaks on one day. The average BC concentration during peak exposure was 4206 ng/m^3. By design no peaks could be encountered in the first 2 h of the day, however we do not expect many peaks in this time window (0.2% and 0.5% of peaks occurred between 2-3 a.m. and 3–4 a.m., respectively). A peak lasted on average 27.5 ± 19.3 min. Peaks were present on 5.5% of the time on average, ranging from no peaks per day to peaks during 23.3% of the time. Although peaks were short and infrequent, BC exposure during peaks contributed to 21.0% of the total exposure during that day (range: 0.0%–83.5%) (see supplemental information).

The short time in transport (8%), was responsible for 32.7% of the peaks; and almost a quarter (24.1%) of the observations in transport were categorized as peak exposure. This number was highest for bike trips (34.0%), slightly lower for trips by car or public transport (27.6%), and lowest for trips on foot (16.6%). Only 0.9% of the exposure while sleeping was peak exposure; during daily activities this was 5.6%. During traffic rush hour (7–10 a.m.; 4–7 p.m.) 12.0% of all observations were categorized as a peak, outside traffic rush hour only 3.4% of the observations were categorized as peak exposure. When we only consider transport activities, 35.3% of the observations in rush hour were categorized as a peak. Likewise, from Monday through Friday participants were more likely to encounter a peak than during the weekend (6.2% on a weekday; 4.1% in the weekend). The chance of encountering peaks differed by season with highest rates in fall, and lowest rates in summer (6.2% in fall; 5.4% in spring; 5.0% in summer; 5.5% in winter). In our sample, the number of peaks per person-day was not related to gender, and the effect of age was close to zero. Consistent results were seen by dataset and study area; with the exception of a lower occurrence of peaks while traveling in London (all travel modes). The number of peaks per person-day was slightly higher in Spain (Barcelona; 3.2 ± 1.6 peaks per day) compared to Belgium (region of Flanders and Antwerp; 2.7 ± 1.6) and the UK (London; 2.7 ± 1.7), the latter caused exclusively by the lower number of peaks while in transport.

The findings from the mixed models that controlled for confounding variables were comparable to the results presented above from the descriptive analysis (see supplemental information). Age was found to be negatively related to the presence of peaks in the mixed model ($p = 0.045$), but the effect was very small (estimate = −0.004378).

Pearson's r correlation coefficient between daily average BC exposure and the number of peaks during that same day was 0.44 (Fig. 2). The association between cumulative daily exposure and cumulative peak exposure during that same day was 0.69. There was an increase in the correlation between average BC exposure and the number of peaks

Table 2
Black carbon concentration during different activities (N = 578,885), and in different transport modes (N = 43,517). Concentrations are in ng/m^3.

Activity type	Black carbon (avg ± SD)	Black carbon (median [IQR])]	Number of 5-min observations
Sleeping	982 ± 898	750 [414, 1268]	175,914 (30%)
Daily activities	1394 ± 2734	874 [478, 1539]	359,454 (62%)
In transport	2656 ± 4186	1390 [641, 3185]	43,517 (8%)
On foot	2085 ± 3925	1121 [559, 2426]	18,413 (42%)
Bike	2736 ± 3784	1733 [743, 3550]	7708 (18%)
Car, Public Transport	3226 ± 4530	1619 [739, 4011]	17,396 (40%)

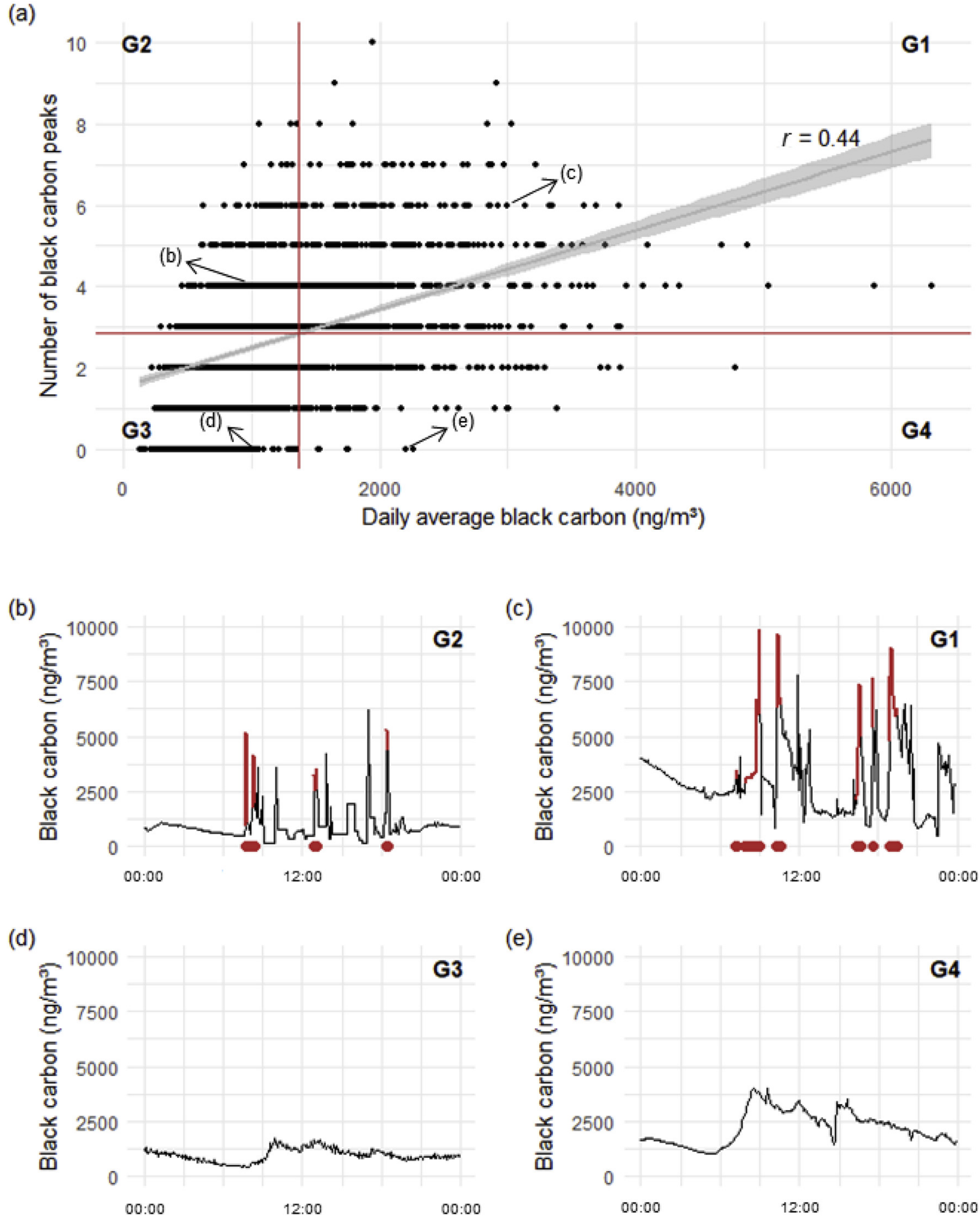

Fig. 2. Correlation plot of daily average black carbon exposure and the number of peaks encountered (panel (a)). The horizontal and vertical red lines on the correlation plot represent the average value that splits the groups (G1 to G4). Four daily timeseries are shown as examples from each group (panels (b)–(e)). The red lines and dots on the timeseries plot indicate the peaks as identified by the algorithm. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

with an increasing number of measurement days per individual, i.e. not considering one person-day (24 h), but up to 18 person-days (Fig. 3). From 10 days onward, the correlation coefficient increased to values above 0.60. However, in longer intervals uncertainties were higher given the lower number of participants with this many measurement days.

We grouped person-days in four groups according to low or high average daily exposure, and a low or high number of peaks on the same day (Fig. 2). As expected, group 1 (high exposure-many peaks) and group 3 (low exposure-few peaks) had the highest number of person-days, respectively 638 and 692. The lowest number of person-days were present in group 4 (high exposure-few peaks): 216. The distribution of the 5-min observations was highly similar in groups 2 & 3 (low exposure-many peaks and low exposure-few peaks), and in groups 1 & 4 (high exposure-many peaks and high exposure-few peaks). There was an association between group and person: a person was more likely to be in the same group over several measurement days. When stratifying the mixed model by group, the determinants of peaks appeared to be

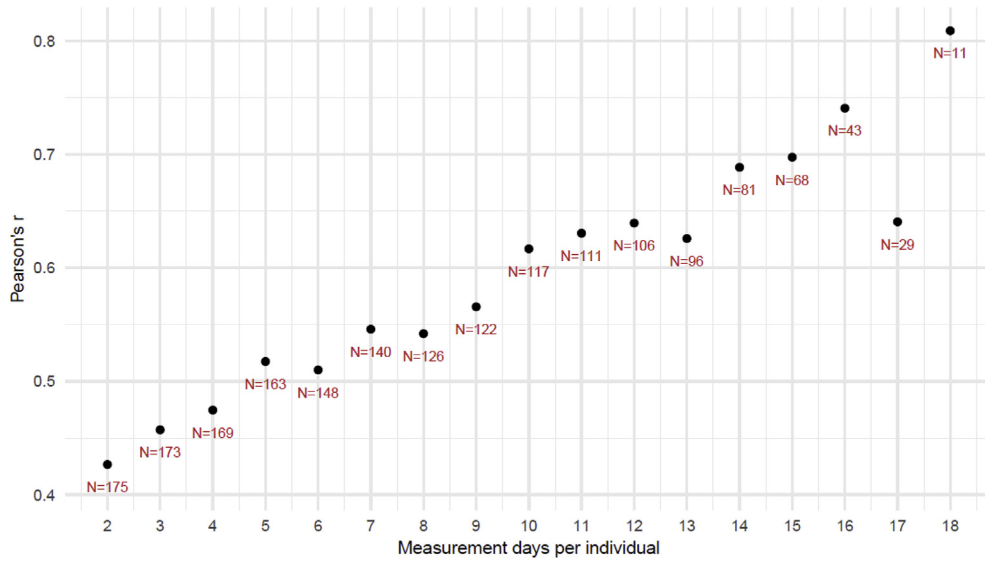

Fig. 3. Pearson's correlation between average BC exposure and the average number of peaks per day, in individuals with up to 18 measurement days (only the first x days per person are considered). The N indicates the number of individuals included in each analysis.

similar in the four groups (see supplemental information).

3.3. Sensitivity analysis

Changing the parameter values of the peak detection algorithm impacted the results in the expected direction (see supplemental information: Tables S2–S4). Lowering the threshold to 1.5 standard deviations increased the frequency (3.7 ± 2.3 peaks per day) and the duration (44.9 min) of peaks. When setting the influence parameter to 1, i.e. a peak is given a weight of 1 in the moving mean, the frequency (1.8 ± 1.2 peaks per day) and duration (14.1 min) of peaks decreased. Shortening the lag to 1 h or 30 min, increased the number of peaks but it had nearly no impact on the average duration of peaks. Changing the parameters did not meaningfully impact the correlation between daily average BC exposure and peak frequency.

By using the median instead of the mean, and the median absolute deviation (MAD) instead of the standard deviation, the peak detection algorithm became less restrictive. The new definition resulted in an average number of 3.0 ± 1.7 peaks per day, and peaks lasted longer to an average of 38.7 min. The correlation coefficient between daily average BC exposure and the number of peaks during that same day became 0.48. Only minor differences were observed in the mixed model results. So, although the model was more sensitive to peaks with the new definition, the general trends and conclusions did not change.

Using a fixed threshold of 2500 ng/m^3 to define a peak, resulted in a higher peak frequency compared to the proposed peak detection algorithm: 3.5 ± 2.5 peaks per day instead of 2.8 ± 1.6 peaks per day. The number of peaks per day ranged from 0 to 16. The average duration of a peak was remarkably longer compared to the proposed peak detection algorithm (42.5 ± 72.1 min versus 27.5 ± 19.3 min), with a maximum duration of 905 min for one peak. The Pearson's r correlation coefficient between daily average BC exposure and the number of peaks during that same day was 0.59 when we used the fixed threshold of 2500 ng/m^3 to define peaks.

4. Discussion

In a large multicenter pooled dataset, we applied a peak detection algorithm to study determinants of BC peak exposure and we investigated the association between average and peak exposure over time. The occurrence of peaks varied substantially across different microenvironments and transport modes. Hour of the day, day of the week, and season were also associated to the presence of peaks. Results were similar in separate datasets. Although peaks were short in duration, our study showed that they made up an important part of daily exposure to BC. However, when we considered a 24-h exposure interval, average BC exposure and the number of peaks showed only a moderate correlation; in longer time intervals (up to 18 days) the correlation coefficient increased.

Previous studies already revealed that daily timeseries of personal monitoring of traffic-related air pollution closely relate to the time-activity pattern of a person (Carvalho et al., 2018; Dons et al., 2011; Koehler et al., 2019; Rivas et al., 2016). A typical exposure pattern of a full-time office worker follows a two-peak pattern: relatively constant concentrations at night, at work, and at home, and two peaks while commuting to and from work. This pattern finds concentrations to be lognormally distributed with a long tail on the right, and by definition this already indicates the presence of peaks (Jeong and Park, 2018). Also, other crude measures as the standard deviation, ratio of mean and median, or the height of the 95th percentile may be indicators of whether and how many peaks are to be expected. But a typical pattern for a full-time worker may not be representative for all situations. Firstly, on non-workdays people have a different time-activity pattern resulting in a different exposure pattern. Secondly, rapidly changing background concentrations for example due to changes in meteorology may lead to changes in exposure independent of the time-activity pattern. And thirdly, some traffic-related air pollutants may have non-traffic sources as well, for example BC from cooking or charbroiling meat (Jeong and Park, 2018; Van Vliet et al., 2013), which will disturb the typical two-peak pattern. In our pooled sample, 2053 unique daily exposure patterns partly representative for the adult population in Europe were analyzed and revealed an average of 2.8 BC peaks per day, and at least part of these peaks could be attributed to time spent in traffic.

About a quarter of the observations in transport were categorized as peak exposure. Most peaks were encountered while cycling, which is in line with findings from Jeong and Park (2018) who also found more BC peaks during active travel compared to traveling by motorized modes. Abraham et al. (2014) found a higher occurrence of peak exposure to BC during walking compared to traveling by bus or on the underground. Bauer et al. (2018) stated that seating location on the bus determined the number of BC peaks. Unfortunately, in our study, because of the inability of the SenseWear armband to discriminate between private and public motorized transport, we could not study peaks in cars and in different types of public transport separately. In a spatial analysis of peak BC exposure while cycling, it was found that peaks were present at (major) intersections, along routes with a high share of heavy duty vehicles, and in a tunnel (Peters et al., 2014). In our study, we found lower concentrations and a lower number of peaks in the transport microenvironment in London. This may be explained by the high number of underground users in London (microAeth may report BC wrongly because of the presence of iron in the underground, however we would expect an overestimation rather than an underestimation), differences in vehicle fleets or fuels (lower share of diesel passenger cars in the UK; share of diesel cars in the total passenger car fleet in 2016: Belgium 60%, Spain 57%, UK 38.7% (EEA, 2018)), or differences in infrastructure and urban design (de Nazelle et al., 2017; Hankey and Marshall, 2015). There was no proof of other differences in peak exposure between cities and countries. The frequency of peaks was lower in the warmer season: background concentrations were lower and potential peaks did not surpass the threshold of 2500 ng/m^3, additionally seasonal variation in the time-activity pattern may have caused a lower occurrence of peaks in the warmer season. The same reasoning explains why there are more peaks on traffic rush hours, and from Monday to Friday.

We found a moderate association ($r = 0.44$) between daily average BC exposure and the daily number of BC peaks, contradicting the hypothesis that the number of peaks and personal exposure are highly correlated and can be used interchangeably. In a sensitivity analysis, we did observe a higher correlation of 0.59 when applying a fixed threshold of 2500 ng/m^3 to define peaks. Epidemiological studies, such as panel studies looking at short term exposure of up to a week, should be aware of the moderate association. In contrast, cohort studies interested in long term exposure, could assume that the number of peaks and longer-term personal exposure are sufficiently correlated, as indicated in our study by an increase in the correlation coefficient when averaging over longer time periods of up to 18 days.

Participants were more likely to be in the same of four groups (grouping determined by average exposure and peak frequency) on multiple days. A similar peak frequency could be explained by participants performing routine activities resulting in similar exposure profiles. Because at least part of the measurement days was on consecutive days with likely similar background concentrations, this resulted in similar average exposures. Within the groups, peaks could be explained by the same factors, as shown in the stratified mixed models.

Limitations of our study include (1) differences in activity reporting between the two studies (electronic diary vs. activity tracker); (2) the 5-min time resolution of the microAeth for BC monitoring; (3) limited generalizability with only data from adults. Activity reporting with diaries used to be a labor-intensive task for participants, but more recently activity trackers such as the SenseWear armband appeared as an alternative requiring minimal intervention from the participant. Unfortunately, both methods have their deficiencies. Self-reporting tools are prone to recall bias, social desirability bias and fatigue effects, and personal characteristics may influence reporting behaviour (Breen et al., 2014). Activity trackers in general, and the SenseWear armband in particular, struggle with activity and transport mode classification. It has been reported that the SenseWear is not as precise in estimating physical activity duration and energy expenditure (SenseWear overestimates) in comparison with a questionnaire and a gold standard method respectively (Laeremans et al., 2017; Santos-Lozano et al., 2017). The difference in activity reporting most probably reduces precision of our results, however, an analysis by dataset did show very similar time-activity patterns pointing to small misclassification errors only. No direct comparison of the electronic diary and the activity tracker (SenseWear) could be made. Secondly, the time-resolution of 5 min prevented us from detecting very short transient peaks in the order of magnitude of seconds (like passing vehicles); though these very short peaks were included in the 5-min averages. This has no impact on our analysis, but it would complicate a spatial analysis of BC hot spots (locations with frequent air pollution peaks). The time resolution was chosen because of practical reasons of internal memory capacity and battery lifetime. Thirdly, both studies included in the pooled sample were on adults only. Children are more susceptible to the health effects of air pollution, and they may have a deviating BC exposure profile because of their different time-activity schedule. However, for adults, we believe we covered a good portion of the variability by including > 2000 person-days of adults living in multiple European countries.

Mobile air pollution sensors enabled us to measure 24-h time-integrated personal exposure, and to assess both peaks and average exposure. This is the first study, to the best of our knowledge, to analyze peak exposure to air pollution on such a scale in volunteers performing their routine activities. Previous research on peak exposure tended to focus on occupational settings only, leading to regulations for many chemicals by setting ceiling limits or short-term exposure limits (Smith, 2001). The ability to delineate peaks in everyday life through space and time allows for correlation of exposure with specific activities, specific microenvironments, or geographic locations (Adams et al., 2009). This could help the design of effective and targeted interventions and policies to reduce average and peak exposure. Further research into the health effects of (repeated) peak exposures is needed, but if it is the upper tail of the exposure distribution that determines the risk, it would be good policy to try to limit the frequency of peaks, rather than only trying to reduce average concentrations (Paoletti et al., 2014). In a study in Slovenia measuring BC in cyclists, the authors found that an alternative and cleaner route led to lower average exposures, but that very short peak exposures, mainly near intersections, were not affected (Jereb et al., 2018).

We propose a generic method to detect peaks in air pollution personal monitoring that on the one hand handles the fact that exposure is inherently variable but that a sufficiently high dose rate is needed to experience acute changes in the body, and on the other hand can deal with peaks of different length and height. For sustained peaks, the algorithm may underestimate the duration of peaks due to the increasing standard deviation. This may also affect the average magnitude of a peak, and the contribution of peak exposures to cumulative exposure. However, with the algorithm we prioritised the estimation of peak frequency and rate of increase, as we believe these variables are most relevant for health. A fixed threshold, for example of 2500 ng/m^3 as tested in the sensitivity analysis, could be useful to study the duration of exposure at high concentrations, but is not very well suited for the detection of events with a high rate of increase of concentrations. On days with high background concentrations this resulted in large parts of the day being labelled as a peak, but excursions above the threshold were not separately marked. The algorithm could be linked to an application that informs participants in real-time about peaks and serve as a warning/alerting system. Moore and colleagues already applied the algorithm described in our paper to detect indoor air pollution peaks in real-time, in combination with ecological momentary assessment through text messaging (Moore et al., 2018). Air pollution alerts could convince the carrier of the device to adapt his or her behaviour, and improve self-management, for example through reducing vigorous outdoor activities or cycling, avoiding air pollution hot spots, reducing automobile use, or altering medication levels (Kelly et al., 2012; Saberian et al., 2017; Ward and Beatty, 2016). Promoting the use of an app or a personal monitor with air pollution alerts should always be combined with emission reduction policies to also protect vulnerable individuals with no access to this technology.

In conclusion, air pollution peak exposures are omnipresent in everyday life with 2.8 peaks per person per day on average. When moving around in a city, one out of four measurements was identified as a concentration peak. Until now the independent impact of repeated peak exposures was not studied, with the main motivation being that the number of peaks and average exposure are highly correlated. In our research we found that this is not always true. In the future, we will link the occurrence of peaks to several cardiovascular markers to test for differences in biological responses to BC pollution from time-varying exposures (with peaks) versus time-invariant exposures. This will help us in gaining a better understanding of the health relevance of repeated peak exposures.

Conflicts of interest

None.

Declaration of interests

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

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.atmosenv.2019.06.035.

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