



Air pollution at human scales in an urban environment: Impact of local environment and vehicles on particle number concentrations

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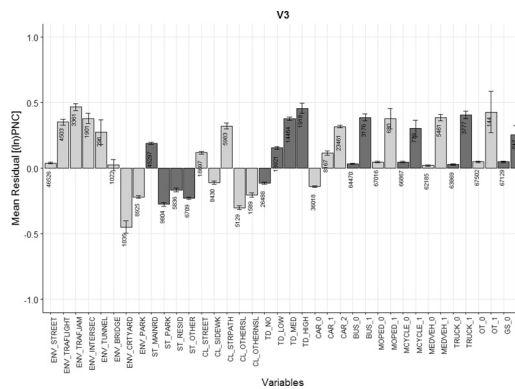
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HIGHLIGHTS

- Differences in ultra-localized air pollution levels were quantified in urban areas.
- Buses, mopeds, trucks showed 30–40% higher than avg. particle number concentrations.
- Parks, green spaces showed 22% lower than average particle number concentrations.
- Avoiding high, mixed vehicle traffic density on main roads would reduce PNC exposure.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 30 January 2019

Received in revised form 18 June 2019

Accepted 20 June 2019

Available online 20 June 2019

Editor: Lidia Morawska

Keywords:

Air pollution

Urban

Exposure

Particle number concentration

Infrastructure

Policy

ABSTRACT

Air pollution is a global challenge causing millions of premature deaths annually. This is limited not only to developing, but also developed nations, with cities in particular struggling to meet air quality limit values to adequately protect human health. Total exposure to air pollution is often disproportionately affected by the relatively short amount of time spent commuting or in the proximity of traffic. In this exploratory work, we conducted measurements of particle number concentrations using a DiscMini by bicycle. Eighteen tracks with accompanying video footage were analyzed and a suite of factors classified and quantified that influence exposure to air pollution. A method was developed to account for variations in the ambient average concentrations per trip that allowed for comparison across all tracks. Large differences in ultra-localized air pollution levels were identified and quantified for factors such as street type, environmental surroundings, and vehicle type. The occurrence of one or more non-passenger car vehicles, including e.g., buses, mopeds, or trucks, result in an increase in particulate concentrations of 30% to 40% relative to the average ambient level. High traffic situations, such as traffic jams or cars waiting at traffic lights, result in increased particulate concentrations (+47% and +35%, respectively). Cycling in residential neighborhoods decreased particulate number concentrations by 17% relative to the ambient average level, and by 22% when cycling through green spaces or parks. Such information is valuable for citizens who may want to reduce their air pollution exposure when moving through a city, but also

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for policy makers and urban planners who make or influence infrastructure decisions, to be able to reduce exposure and better protect human health, while progress is made to reduce air pollution levels overall.

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1. Introduction

Air pollution is currently one of the foremost environmental health problems. The Global Burden of Disease Study estimated that 4.2 million premature deaths resulted from outdoor air pollution in 2015 (Landrigan et al., 2018). The adverse health implications from air pollution exposure are particularly critical in urban areas, which are population centers, but also hotspots for emissions, in particular those originating from traffic. In Europe, 82–85% of the urban population was exposed to ambient air pollution concentrations of PM_{2.5} (particulate matter with an aerodynamic diameter of 2.5 µm or less) that exceed the World Health Organization (WHO) guidelines from 2013 to 2015 (EEA, 2017). Exposure and health effects are often assessed using air pollution data from fixed urban background monitoring stations. A number of studies have shown however, that such monitoring station data leads to significant underestimates of the exposure of population subgroups, or in some cases shows little to no association with exposure at all (de Nazelle et al., 2017; Gulliver and Briggs, 2004; Kaur et al., 2007; Ragettli et al., 2013), indicating the need for investigation of more realistic levels of pollutant exposures. Furthermore, the larger size fractions of PM, such as PM_{2.5} and PM₁₀ (particulate matter with an aerodynamic diameter of 10 µm or less) have been found to be poor indicators of traffic emissions for studies examining exposure (Boogaard et al., 2009; Kingham et al., 2013), while a number of studies all consistently found that particle number concentration (PNC) or ultrafine particles (UFP) are far better indicators for local traffic-related air pollution than PM_{2.5} or PM₁₀ (Boogaard et al., 2009; Kaur et al., 2007; Kingham et al., 2013). Despite this limitation, far more studies evaluating differences in commuting environments/transport modes have looked at PM mass concentrations rather than PNC (Karanasiou et al., 2014). Evidence also indicates that exposure to UFP (the dominant fraction contributing to PNC) is related to significant adverse health effects (Delfino et al., 2005). Finally, a significant part of air pollution exposure can occur during commuting or time spent in transit through cities. Specifically, elevated levels of pollutants in transport microenvironments may contribute significantly to daily total exposure despite the short period of time (Chaney et al., 2017; Karanasiou et al., 2014; Kaur et al., 2007; Michaels and Kleinman, 2000; Ragettli et al., 2013; Weichenthal et al., 2011). For example, Dons et al. (2012) found that while only 6% of time was spent in transport, it accounted for 21% of personal exposure to black carbon and ca. 30% of inhaled dose. Associations of in-traffic exposure to particle number and soot to changes in lung function have also been found (Zuurbier et al., 2011).

A growing number of studies look at exposure to air pollutants and the variation between different transit modes (e.g., car, bus, bicycle, walking), including the difference in exposure between direct and alternative routes. There are substantial differences in the findings among these studies. For example, some studies found that commuters in motorized modes (car, bus, and/or motorcycle) experienced higher exposure to pollutants such as PM_{2.5}, black carbon, and PNC relative to pedestrians or cyclists (Kingham et al., 2013; Morales Betancourt et al., 2017), while other studies identified higher exposure to the same pollutants while cycling relative to driving (Good et al., 2016; Okokon et al., 2017). While other studies found a mix, with generally the highest exposure for the car transport mode, and the lowest for pedestrians, with bus and bicycle transport modes in the middle of the exposure range for PM_{2.5} and UFP (Kaur et al., 2007). In some cases, the exposure level was found to be pollutant dependent (Good et al.,

2016). These differences show a need for greater understanding of what factors influence the differences in exposure that are being observed.

Many of these studies, however, consider only exposure and not dose. And despite lower exposure, results showed that a higher dose can result owing to higher inhalation rates for active transport modes such as walking or cycling (Morales Betancourt et al., 2017). This result is consistent across a number of studies, including those that identified higher exposure for car drivers or bus passengers, emphasizing the importance of considering inhalation rates (de Nazelle et al., 2012; Int Panis et al., 2010; Ramos et al., 2016). However, studies have also shown that the beneficial effects of increased physical activity far outweigh the potential mortality effect of increased air pollution doses and traffic accidents (de Hartog et al., 2010; Tainio et al., 2016). Additional societal benefits owing to the reductions in air pollution, greenhouse gas emissions and traffic accidents were also found. The majority of these studies emphasize the need for separation of cyclists from motorized vehicle traffic, or at minimum, routes that facilitate reduced traffic volume, to foster a beneficial shift in mobility and reduction in exposure.

A number of studies have also evaluated the differences in route and the implications for cycling exposure. Overall, these studies found that taking alternative routes to avoid main roads, riding off-road rather than on-road, or in bicycle lanes on sidewalks rather than mixed traffic lanes, all result in reduced exposure to air pollutants such as UFP, PM, BC and CO (Good et al., 2016; Kingham et al., 2013; Morales Betancourt et al., 2017; Ragettli et al., 2013; Thai et al., 2008). The study by Good et al. (2016) however, also noted that while alternative cycling routes with reduced traffic may result in lower exposure of some pollutants, many multi-use cycle paths (those paths that are not contiguous with a roadway) that would likely result in more substantial reductions in exposure, are not practical for many users (e.g., too out of the way, extends the trip distance). Such considerations are important when designing cycle infrastructure and considering the implications for exposure.

While it is important that further studies are conducted to better understand exposure to air pollution across transit modes, studies that investigate the factors that influence the air pollutant concentration a person is exposed to during transit are far more limited – typically to a characterization of traffic density (Zagury et al., 2000; Zhu et al., 2002). In a review paper by Kaur et al. (2007), factors influencing personal exposure concentrations in the urban transport microenvironment were discussed, such as distance from roadways, traffic density, and meteorological conditions. Findings showed that gradients in pollution (vertical and horizontal) resulted in lower concentrations of air pollutants as the distance from the roadside increased. Meteorological influences, such as increased wind speed resulted in decreased exposure, and wind direction will affect the sources influencing a location (Kaur et al., 2007; Thai et al., 2008). A handful of studies were highlighted that observed relationships between traffic count or traffic density and particle number concentrations (Kaur et al., 2006, 2007; Zhu et al., 2002 and references therein). In one of the few studies to quantitatively assess predictor variables' associations with particle number concentration, Boogaard et al. (2009) identified passing vehicles, waiting for traffic lights, passing through intersections, and bicycle lanes/paths close to motorized traffic as factors that significantly predicted PNC variability during cycling in a study across 11 cities in the Netherlands.

In this exploratory study our research questions were (1) what factors influence particle number concentrations that we are exposed to? (2) can we quantify the amount of influence such factors have on exposure during transit through urban environments using bicycle measurements? To address these questions we developed new methods for application to this dataset of diverse bicycle routes to systematically and quantitatively evaluate the variables that lead to higher or lower levels of particle number concentration, in a manner that is directly relevant to exposure in urban environments. This study includes variables such as amount of traffic, but goes beyond to evaluate variables such as road type, presence of different vehicle types, and local environment, among others. Such information is valuable for citizens who may want to reduce their exposure when moving through a city, but also for city governments and others who make or influence infrastructure decisions, to be able to reduce exposure and better protect human health, while progress is made to reduce air pollution levels overall.

2. Methods

The data were collected in the metropolitan area of Berlin and Potsdam in the context of a larger measurement campaign including stationary and mobile measurements carried out in the summer (June through August) of 2014 (Bonn et al., 2016; von Schneidmesser et al., 2018). Measurements of particle number concentration were collected during bicycle trips, many of which were morning or evening commutes on commonly used cycling routes through the two cities. In addition to the PNC measurements, video documentation of the routes was collected, which allowed for evaluation and identification of the suite of variables. The unique combination of information allowed for a number of questions to be addressed including the importance of local areas (park, residential neighborhoods, etc.) and the proximity to specific potential sources of pollution including buses, trucks, and cars. Techniques were developed to analyze the data which included video analysis and statistical methods to identify average ambient concentrations. Variations in these analytical methods were applied to assess the robustness of the results.

2.1. Particle number concentration measurements

Particle number concentration measurements were collected using a DiSCmini (Matter Aerosol AG, Switzerland). The DiSCmini is designed to detect particle number concentrations using a charged equilibrium in the size range of 10–300 nm (modal diameter) with an impactor for particle size cut off at ca. 700 nm, and is capable of capturing a linear signal for the concentration range between 10^3 and 10^6 particles cm^{-3} . For further instrument details, please see Kaminski et al. (2013). Studies comparing the DiSCmini to condensation particle counter (CPC) instruments have shown good agreement and indicated that the DiSCmini can appropriately be used for personal exposure studies (Kaminski et al., 2013; Meier et al., 2013; Mills et al., 2013; Viana et al., 2015). This comparison includes similar deployment set-ups as in this work, on mobile platforms in outdoor environments including traffic environments, where the data from a DiSCmini and two CPC instruments were found to be highly correlated for particle number and average particle size, with differences in total particle counts attributed to the difference in size ranges that were instrument specific (Meier et al., 2013). For example, a field comparison showed good agreement (r^2 values generally over 0.8) and 10–18% relative mean difference for PNC between a DiSCmini and a CPC (Viana et al., 2015). The DiSCmini was placed in a backpack (on occasion a bike pannier) with a tube extending out of the bag to provide the inlet for sampling ambient air. The sampling height therefore depended on the bicycle and cyclist's set-up, but was generally 1.0 ± 0.25 m above the ground. Previous work found that the vibrations owing to the implementation of the instrument while cycling did not have an effect on the measurement coefficient of variation (Gerwig and Wirtz, 2015). An inter-comparison experiment using

ambient air sampling was carried out two months prior to the start of the campaign, where the DiSCmini was compared to a TSI CPC Model 3772. This DiSCmini showed good agreement with the CPC with an r^2 of 0.99. Furthermore, performance checks were carried out by comparing the DiSCmini to a TSI CPC Model 3776 directly before and after the measurement campaign, and to a TSI NSAM 3550 before, midway, and after the measurement campaign. These checks (carried out between 22.5.2014 and 21.5.2015) showed good comparability between the DiSCmini and the TSI NSAM 3550 and CPC 3776 of 82–100% (average $87\% \pm 9\%$ SD, $n = 7$) and 56–76% (average $65\% \pm 12\%$ SD, $n = 6$), respectively. The differences in particle size detection limit for the CPC 3776 ($D_p > 2.5$ nm), NSAM 3550 ($10 < D_p < 1000$ nm), and DiSCmini ($10 < D_p < 700$ nm) are reflected in the comparability numbers. The particulate data were evaluated to determine their statistical behavior. QQ-plots showed that the data behaved in a log-normal manner and thus data were natural logged before using them in the study to assure high concentrations did not have an excessive impact on the conclusions. The data were also tested for autocorrelation, and, as expected showed autocorrelation for both 1 s data (original resolution) and 30 s averages (e.g., Fig. S3); the effect of autocorrelation was included in the estimates of standard error on means and differences. Data from all tracks showed a mean particle number concentration of 7680 particles cm^{-3} (median 7020 particles cm^{-3}) with a standard deviation range from 3260 to 18,070 particles cm^{-3} based on the log-normalized data.

All bicycle routes covered in this study are shown in Fig. 1. The routes shown were part of a larger set of mobile measurements including further cycling measurements, as well as measurements by van, air plane and glider as depicted in Bonn et al. (2016). The bicycle routes included a variety of environments and road types, that ranged from wide, paved cycle paths that traversed through forested areas, to cycle paths that either shared a bus lane, were part of the road but as a separate bike lane, or shared the sidewalk, typical constellations for cycle paths on main roads with higher traffic, to routes through residential streets with lower traffic where no specific cycle path is designated, and routes that included urban parks, often on gravel or otherwise unpaved paths.

2.2. Cycle routes and video data classification

During the mobile bicycle measurements a Garmin VIRB Elite HD Action Camera with GPS was enabled to capture the route, environmental surroundings, and emissions sources (mainly cars, trucks, buses, and other traffic influences) that could influence the particle measurements. The camera was mounted on the cyclist's handlebars. Video data from approximately one month of cycling was analyzed at one second resolution. In total, 18 cycle tracks were included in the analysis. The 18 cycle tracks took place between 11 June and 3 July 2014, on weekdays, with the exception of one track on a Saturday. The time of day for the tracks was between 7:00 and 10:30 (local time) for 7 tracks, between 16:30 and 21:30 for 9 tracks, and between 13:00 and 14:00 for 2 tracks, one of which was the track on Saturday. Routes were not prescribed, but typically followed commuting routes of the cyclists. Cycle track duration ranged from 45 min to 3 h, with an average duration and standard deviation of 1 h 31 min \pm 34 min. This corresponded to tracks that ranged from 7.7 to 35 km in length (23 ± 9.3 km). In some cases, owing to instrument or video malfunction, data for the entire duration of the track were not available. These tracks are not included in this study. The DiSCmini measurements and video evaluation were matched via time stamp that was checked prior to each cycling route to ensure consistent time stamps.

The video data were evaluated for a variety of variables that could potentially influence the particle number concentrations. The following variable categories were evaluated: presence of the number of different types of vehicles (referred to as the 'event' variables), traffic density, environment, street type, and cycling location. The event category included eight variables, such as passenger car, bus, moped, etc. and was



Fig. 1. Map of all cycle track routes included in the analysis presented here.

classified by 'occurrence' or counts that reflected the number of vehicles in the frame. The environment category included eight variables to provide detailed information as to the environment the cyclist was passing

through, whether this was e.g., an on road cycle lane, stopped at a traffic light, or passing through a tunnel. Further details to variable classifications in each category are provided in Table 1.

Table 1
Video analysis variable categories.

Variable	Category code	Category description
Environment (ENV)	ENV_STREET	On street/road, whether marked as a cycle path or not, not size dependent, includes small crossings (only when light is green)
	ENV_TRAFLIGHT	Waiting when traffic lights are red
	ENV_TRAFJAM	Passing by/in a traffic jam, includes approaching red traffic lights with waiting vehicles
	ENV_INTERSEC	Crossing an intersection
	ENV_TUNNEL	Cycling through a tunnel, underpass or under a bridge
	ENV_BRIDGE	Crossing a bridge (over water or another road)
	ENV_CRTYARD	Coming out of or going into a courtyard/backyard/parking area
	ENV_PARK	Cycling in park/larger green space not directly next to a road
Street type (ST)	ST_MAINRD	Urban street/road, including multi-lane thoroughfares
	ST_PARK	Cycle road through a park/larger green space (mainly Kronprinzessinnenweg)
	ST_RESID	Cobblestone (smaller) streets or otherwise residential streets
	ST_OTHER	Park, parking, pedestrian, or backyard, (not a roadway)
Cycling Location (CL)	CL_STREET	On street (no cycle path marked)
	CL_SIDEWK	On the sidewalk (sealed surface)
	CL_STRPATH	Marked cycle path on the street
	CL_OTHERSL	Path not on roadside, sealed surface
Traffic density (TD)	CL_OTHENSL	Other (e.g. dirt roads, with unsealed surface and gravel)
	TD_NO	No traffic
	TD_LOW	Low traffic: at least 1 vehicle in picture, vehicles pass sporadically; one or two vehicles have passed or almost passed out of the picture before the next one appears
	TD_MED	Medium traffic: ca. 3–5 vehicles; vehicles pass by at a higher frequency
	TD_HIGH	High traffic: 5 or more vehicles; vehicles pass close behind each other, continuous dense traffic flow
Events (occurrence of vehicle types)	CAR	Based on the presence of these vehicle types in the video frame; the following categories exist: CAR_0, indicating the absence of cars; CAR_1 indicating the presence of 1 car; CAR_2 indicating the presence of 2 or more cars
	BUS	Based on the presence of these vehicle types in the video frame; for all variables the categories e.g., BUS_0 and BUS_1 exist, indicating that there are no buses present or that there are 1 or more buses present. *Vintage or older vehicle, in Berlin this can include old Trabant vehicles that are used for tours, generally would expect this to correspond to higher emissions
	MOPED	
	MCYCLE	
	(motorcycle)	
	MEDVEH (medium vehicle)	
	TRUCK	
	(semi-truck)	
	OT ('old timer')*	
	GS (gas station)	

2.3. Data analysis

Owing to the time intensity of the associated video data classification, only a subset of the cycle tracks (18 tracks of 85 total tracks) from the three month campaign were analyzed and included in this study. To ensure enough data points across the event variable categories, such as bus, truck, moped, etc. higher occurrence classifications were combined. For example, as shown in Fig. S1, there were 2887 counts for BUS_1 (indicating the presence of 1 bus). However, for BUS_2 (indicating 2 buses) or BUS_3 (indicating 3 or more buses), there were only 271 and 16 counts, respectively. These higher occurrence classifications (e.g., BUS_2 and higher) were combined with BUS_1, to simply indicate whether or not buses were present in the video frame, as the amount of occurrences in the event variable categories of >1 were typically limited. For the bus case, considering only occurrence categories 1 through 4 (excluding BUS_0 where no buses were present), categories 2 through 4 were only 9%, of which count 2 contributed 8.5%. For a number of the other categories, counts of 2 or more were even fewer (see Fig. S1 for all occurrences by variable category). Among the environment variable, the STREET classification was the most common by a good margin, with 69% of the total counts. The second most common was PARK with 13% of the total counts. This is consistent with the street type classifications, with the largest classification being main roads (67%), 14% of the total counts streets passing through parks, and 8.6% residential streets (Fig. S1).

Video classification data for the variables were matched to particle number concentration data using two different approaches to understand the temporal matching of potential particle source information and particle concentrations. The match options were (1) a straightforward match of video analysis and measurement by time stamp at the original one second resolution, (2) an average of the particle number concentration data to an interval of 30 s, with the time stamp reflecting the mid-point of the averaging time, matched to the highest classification for occurrence-based video category classifications (e.g., traffic intensity), or the most frequently occurring classification for other variables (e.g., environment) over the associated 30 s time period. The two approaches were used to assess whether there was an offset between the classification information from the video and the particle concentrations measured. The 30 s time period was chosen to have sufficient length to account for a possible offset for particle concentrations being measured before or after their emissions source was visible in the video, while not being so long in duration that too much distance was covered and too many different classifications included. Match option 1, with the highest temporal information, was considered the primary assessment method.

For each approach, the video data were evaluated for frequency counts for each variable and the corresponding categories (see SI Fig. S1), and were compared with the particle number concentration by variable category using box and whisker plots (Fig. 2), prior to log-normalization.

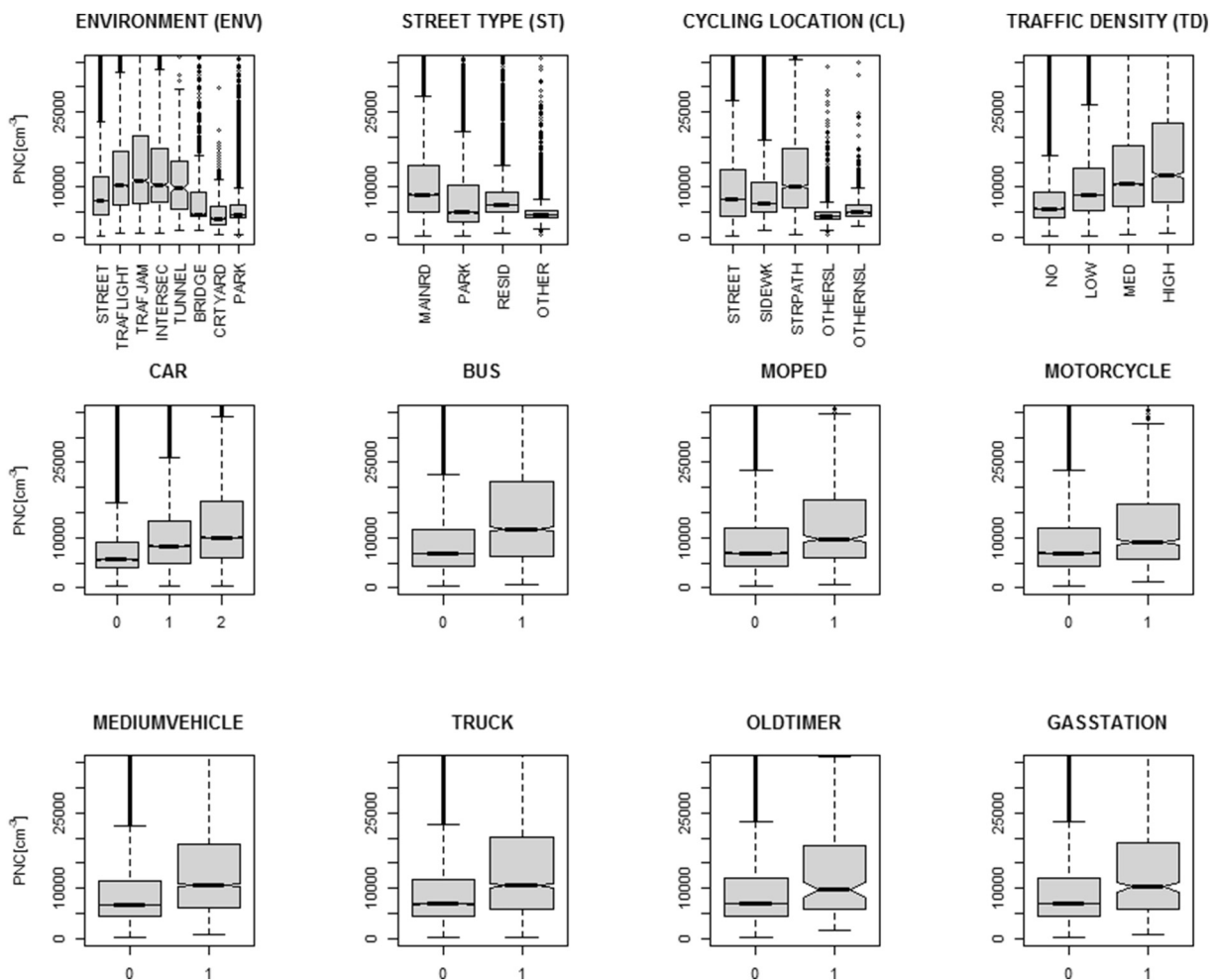


Fig. 2. Box and whisker plot of particle number concentrations by variable category classification, prior to log-transforming the data. The lower and upper bounds of the box correspond to the first and third quartiles of the data, the upper and lower limits of the whiskers extends to $1.5 \times \text{IQR}$ (inter-quartile range), all points beyond those are outliers. The notches around the median extend $1.58 \times \text{IQR} / \sqrt{n}$ and indicate a ca. 95% confidence interval around the median. The y-axis scale shown is consistent across all variable categories and is limited so that the confidence intervals around the median are sufficiently visible.

To account appropriately for ambient conditions, we evaluated different methods for establishing average ambient concentrations – a type of background – for each track. This facilitated comparisons of data across tracks, and was needed to account for the influence of meteorology, which would have a strong influence on the particle number concentration measured on any particular day, as well as general differences in the regional background level of particulate air pollution. The primary method for establishing the average ambient level (AA1) for each track was based on the mean particle concentration measured under the environment variable category ENV_STREET (on street/road/cycle path), with traffic density (TD) levels of 0 or 1 (no (TD_NO) or low (TD_LOW) traffic), see Fig. S4. Previous work has showed that mobile measurements at locations with low traffic intensity are useful for the determination of the background concentrations (Van Poppel et al., 2013). In this case, a stationary background site with equivalent PNC measurements was not available for background correction. Furthermore, given the differences in location of the routes, a more local background correction was found to be most appropriate. Three options were tested for background correction, including a mean of all the data, ENV_STREET, and ENV_STREET + TD_NO + TD_LOW. While the options for background correction were quite similar (see Fig. S10), the choice of ENV_STREET + TD_NO + TD_LOW was chosen as the option most similar to urban background. Furthermore, we chose not to use a mean of all the data as the amount of data points in e.g., high traffic environments or low traffic environment (parks) varied from route to route and could result in an over- or underestimation in the background correction. The ENV_STREET + TD_NO + TD_LOW mean value was then subtracted from the data for that track and the residuals were used for completing the analysis. This allowed for the assessment of the effect of e.g., highly trafficked conditions or the presence of buses, relative to the average ambient level across all tracks that were analyzed. We refer to this value as the average ambient level rather than e.g., urban background, as these were determined for each track and to distinguish them from the urban background levels measured at stationary monitoring sites within the city as part of the regulatory air quality network. A meaningful comparison to the urban background concentrations measured in the regulatory network is challenging and beyond the scope of this paper owing to the difference in instrumentation, size fraction, and unit (mass vs PN) measured. The residuals were then combined across all tracks and evaluated. An alternative method for determination of the average ambient level (AA2) was also evaluated, which used a loess-fit (span = 0.5) to the environment variable category ENV_STREET data only (Fig. S4) which allowed us to account for the variation in ambient conditions throughout the duration of a track more explicitly.

3. Results

All 18 cycle tracks included in the analysis, showed a median particle number concentration of 7020 particles cm^{-3} with a standard deviation around the mean from 3260 to 18,070 particles cm^{-3} based on the log-normalized data. By comparison, the median PNC for all tracks, including those for which video evaluation was not carried out, was 7480 particles cm^{-3} with a standard deviation around the mean from 3020 to 15,880 particles cm^{-3} , indicating consistency in the dataset. The overall particle number concentration for all tracks and all variable classifications before the natural log application and any average ambient concentration subtraction are summarized in Fig. 2. The patterns in the box and whisker plots show reasonable relationships between particle number concentration and e.g., event variables such as the number of cars, buses, and trucks, as well as for traffic density, where concentrations increased as the number of vehicles increased. The relationships identified are robust in that they hold regardless of matching method (see Fig. S2 for the 30 s average match, compared to Fig. 2 for the direct time match). Furthermore, the notches on the box and whisker plots indicate a 95% confidence interval for comparing the median values. That

is, if the extent of the notches around the median does not overlap from one variable category to the next, this is an indication of a statistically robust difference in terms of the median concentrations between the categories compared. For example, the median concentration measured in the case of no buses (BUS_0) is shown to be statistically significantly lower than those cases where one or two buses (BUS_1, BUS_2) were identified as present. The variable categories with fewer occurrences have much larger confidence intervals around the median. For variable categories within e.g., environment, the categories are not ordered; there is no increasing or decreasing relationship that evolves from the first category to the last that would be expected. There are however, some statistically significant differences in median values observed between e.g., those categories related to green spaces (ENV_CRTYARD or ENV_PARK) and those related to high traffic conditions (ENV_TRAFLIGHT, ENV_TRAFJAM). The large number of high PNC values beyond the top of the whiskers in these plots (outliers) is indicative of the log-normal distribution that has yet to be accounted for in the Figure.

Analysis evaluating the relationship between street type and the event variables was also carried out. These results show that of the total street type data points collected, two thirds (67%) were identified as main roads; among the street type data, the occurrence of 1 or more buses, mopeds, motorcycles, medium vehicles, and trucks is largely only on main roads (87–97%). Considering only the data points in one street type category, there were negligible occurrences (<1%) of all event occurrences for residential streets, except medium vehicles which were 1.3% of counts, and 1 or more cars, which were 10% of counts. Of the total counts for parks (ST_PARK), only 2.5% of counts were 1 or more buses, 3.3% were medium vehicles, and 4.9% were trucks. Among the counts for main roads (ST_MAINRD), cars, buses, medium vehicles, and trucks were present for 64%, 6.4%, 11%, and 7.2% of data points. The occurrences in parks are owing to the fact that a number of cycle routes that pass through small neighborhood parks are just inside the park boundary along a street, or because of streets that intersect with bicycle routes going through larger parks that are not directly alongside a street. In addition, there were occurrences of maintenance vehicles (medium vehicles or trucks) for a number of the tracks that included a frequently used bicycle path through a larger forested area between Potsdam and Berlin. For more information, see the histograms in Fig. S8.

3.1. Residuals by variable categories relative to the ambient average level

Results presented here reflect the PNC data that were matched with video data evaluated for the suite of variables, such as presence of different vehicle types, traffic density, environment, etc. To compare across routes and days an ambient average PNC for each route and day was determined and subtracted. Variations in the methodology were assessed and found to be robust. For more details, see the methods section.

Fig. 3 shows the results from the variable categories of the particle number concentration residuals (i.e. after average ambient concentration subtraction and application of the natural log). The residuals are presented with error bars representing 1.96 times the standard error, the 95% confidence intervals of the data, indicating statistically significant differences where these do not overlap with zero or among the different categories. All values including confidence intervals and n-values are summarized in Table 2. Among the event variables, it is shown that relative to zero cars (CAR_0), which is significantly below the average ambient level (−14%), the presence of cars (CAR_1 or CAR_2), indicates an increase in the amount of particle pollution measured at each step, for CAR_1 (12%) and CAR_2 (32%) as indicated by their positive residuals. The average ambient level was calculated based on the environment variable classification ENV_STREET, with the traffic density variable including TD_NO or

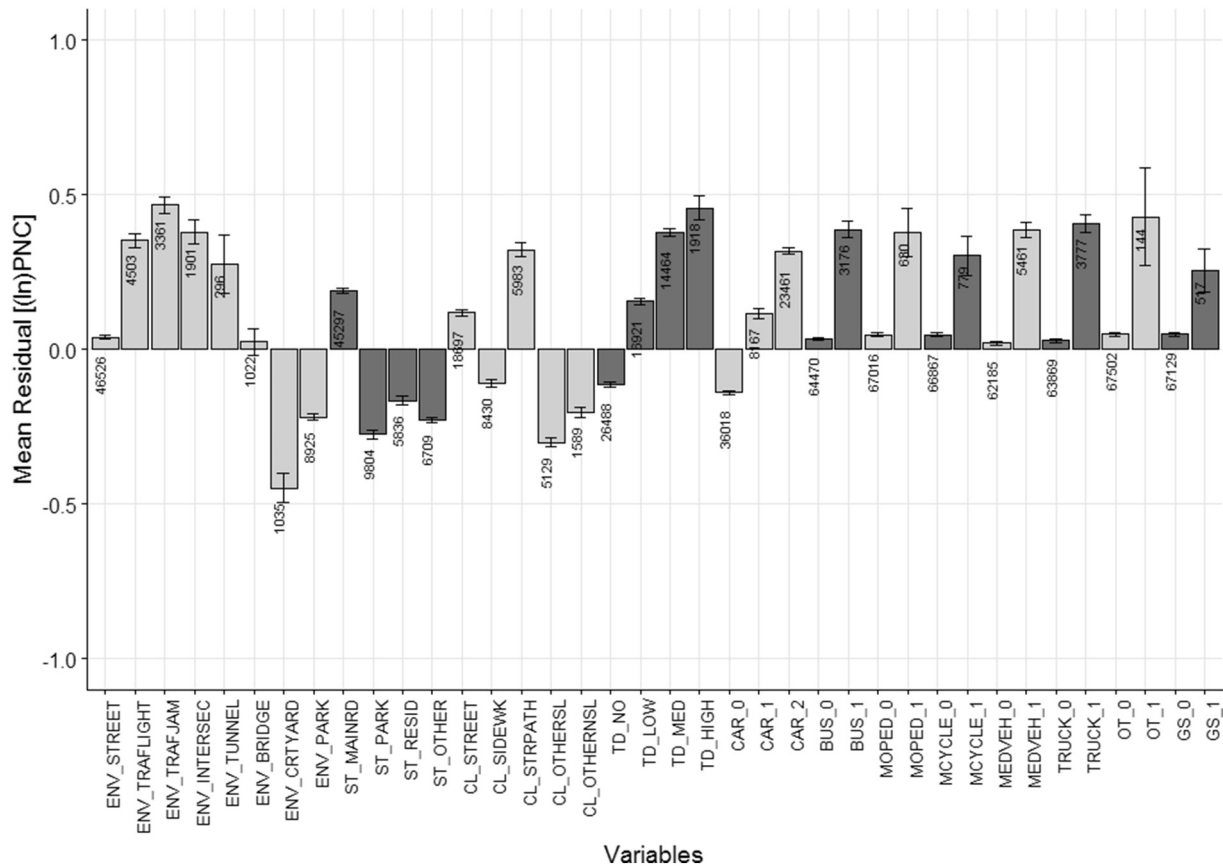


Fig. 3. Mean log particle number concentration residuals by variable category for all tracks after subtraction of the ambient average. Results are from the primary method (direct video matching and AA1). Residuals indicate a change in terms of percent difference from the ambient average concentration. Error bars indicate the 95% confidence interval of the mean.

TD_LOW (no or low traffic) in an effort to capture an average ambient air pollution level for each cycle track.

The same pattern is true for buses, medium-sized vehicles (smaller trucks), mopeds, motorcycles, and trucks (larger trucks, e.g., semi-trucks), with a significant increase in the amount of particles measured in the presence of one or more of these vehicles; the increase relative to the average ambient level ranged from 30% to 41%. Furthermore, the increase tends to be of a greater magnitude than that of CAR_1, and in some cases also more than CAR_2. As most of the values indicating presence originate from only one e.g., bus or truck, being identified in the video ($\geq 86\%$), this indicates that the particulate pollution originating from these types of vehicles tends to be higher than that of passenger cars. Finally, a similar pattern is also observed for the traffic density variable. Where no traffic was observed (TD_NO), the residuals are 11% below the average ambient level. For all other traffic densities (TD_LOW, TD_MED, TD_HIGH), the results show significant increases in the calculated residuals, increasing from 16% to 46% above the average ambient level. These results are qualitatively logical; however, this study allows the quantification of the impact of higher traffic on pollution experienced by pedestrians and bicyclists due to increased traffic.

In terms of the environment variable, the residuals are only slightly (4%) above the average ambient concentrations for cycling on the street or path along a road (ENV_STREET). Given that this was part of the average ambient determination criteria, but limited to low or no traffic conditions, this result is reasonable and reflects the presence of the higher traffic conditions that were included for this residual calculation. In comparison, the data where the cyclist was approaching or stopped at traffic lights, in or passing a traffic jam (ENV_TRAFLIGHT and ENV_TRAFJAM), as well as crossing a street (ENV_INTERSEC) or cycling through a tunnel (ENV_TUNNEL), all show elevated particle number

concentrations relative to the average ambient level, with increases of 28% to 47% (Table 2). In contrast, those data where the environment variable was related to vegetated areas, parks/courtyards/backyards (ENV_CRTYRD) and parks/green spaces (ENV_PARK), showed decreases in PNC relative to the average ambient of -45% and -22% , respectively.

The residuals for the street type variable showed higher than average ambient particle number concentrations for the categories of urban streets, which included larger, multi-lane streets within the city (ST_MAINRD, 19%), and lower particle number concentrations for residential streets (including cobblestone streets which generally include only non-main roads) (ST_RESID, -17%) relative to the average ambient. In addition, for the cases where the cyclist was no longer on a road or path, but rather in a park, in a pedestrian area, or backyard (ST_OTHER) residuals below the average ambient were quantified (-23%), which is comparable to ENV_PARK (-22%) and ST_PARK (-27%). Finally, in terms of the cycling location variable, cycling on the street (the cyclist is part of vehicle traffic, either on main or residential streets where no cycle infrastructure is marked: CL_STREET, 12%) or on marked on-street cycle paths (CL_STRPATH, 32%) led to positive residuals, while cycle paths that were part of a shared sidewalk (CL_SIDEWK, -11%), as well as those not roadside with either a sealed surface (CL_OTHERSL, -30%) or non-paved paths (CL_OTHERNSL, -20%), showed residuals below the average ambient level. We hypothesize that the greater residual observed for the marked on-street cycle path (CL_STRPATH) relative to the lower residual observed for cycling on the street without any markings (CL_STREET) is related to the size of the street and associated traffic density, where those streets with marked cycle paths are more highly trafficked.

Table 2
Residuals relative to the estimated ambient average over all tracks by variable category. Data shown are for the primary methods applied (direct matching and AA1).

Variable category	N	Mean residual (%)	95% C.I. (%)
ENV_STREET	46,526	4.0	0.70
ENV_TRAFLIGHT	4503	35	2.2
ENV_TRAFJAM	3361	47	2.9
ENV_INTERSEC	1901	38	3.8
ENV_TUNNEL	296	28	9.4
ENV_BRIDGE	1022	2.5	4.4
ENV_CRTYARD	1035	−45	4.8
ENV_PARK	8925	−22	0.88
ST_MAINRD	45,297	19	0.73
ST_PARK	9804	−27	1.4
ST_RESID	5836	−17	1.4
ST_OTHER	6709	−23	0.82
CL_STREET	18,697	12	1.1
CL_SIDEWK	8430	−11	1.2
CL_STRPATH	5983	32	2.2
CL_OTHERSL	5129	−30	1.4
CL_OTHERNSL	1589	−20	1.7
TD_NO	26,488	−11	0.73
TD_LOW	16,921	16	1.2
TD_MED	14,464	38	1.3
TD_HIGH	1918	46	3.8
CAR_0	36,018	−14	0.68
CAR_1	8167	12	1.7
CAR_2	23,461	32	1.0
BUS_0	64,470	3.4	0.58
BUS_1	3176	39	2.7
MOPED_0	67,016	4.8	0.57
MOPED_1	630	38	7.7
MCYCLE_0	66,867	4.8	0.57
MCYCLE_1	779	30	6.2
MEDVEH_0	62,185	2.1	0.58
MEDVEH_1	5461	39	2.4
TRUCK_0	63,869	3.0	0.58
TRUCK_1	3777	41	2.9
OT_0	67,502	5.0	0.57
OT_1	144	43	16
GS_0	67,129	4.9	0.57
GS_1	517	26	6.9

3.2. Robustness of results

While the results discussed above (and presented in Fig. 3) are from the application of the direct match video-data matching method, an additional matching method was also evaluated, whereby 30 s average data was used. Furthermore, an alternative background subtraction method (loess fit) was also evaluated (AA2). Overall, the different choices in both matching method and background subtraction method show results that are very consistent with those already presented. While small differences in the magnitude of the residual results exist, owing to the application of the various method choices, the overall patterns and main messages from the results remain consistent. For a detailed discussion of the evaluation of the variations in methodology, please see Text S1.

In addition, given that the routes were not prescribed and occurred at different times of day, we did an analysis to compare the results of the primary method (direct match, AA1) for subsets of the data that included only routes during the morning hours (7:00 and 10:30; 7 tracks) to those during evening hours (16:30 and 21:30; 9 tracks). While some variation in the magnitude of the residuals was observed, the overall patterns remained consistent. For example, the residuals for the traffic density variable classifications, TD_NO and TD_LOW, used for the ambient average determination were within 3% of each other for morning and evening tracks. Other variable classifications, such as those for cycling location, showed somewhat larger differences, e.g., CL_SIDEWK and CL_STRPATH had residuals of −14% and 38% for the morning tracks and −8.4% and 28% for the evening tracks, respectively. These do not

however, change the overall message for the variables. For a comprehensive comparison, see Fig. S7 and Table S2.

4. Discussion

This exploratory work is one of the first studies (and the first in Germany) that investigated the differences in environmental variables affecting the PNC that a cyclist is exposed to quantitatively. Many of these results confirm what we may intuitively hypothesize, e.g., higher traffic density leads to higher PNC or cycling through green spaces reduces the PNC that a cyclist is exposed to; but show surprisingly large variations due to very local factors. For example, the presence of one or more buses, mopeds, or trucks leads to increases in PNC of >30% relative to the ambient average. Factors such as traffic density and the event variables are mutually reinforcing, more so than street type and the event variables which are also related. An evaluation of the relative frequency of event occurrences by traffic density classification shows that, considering the total number of counts within a traffic density category, there tends to be an increasing presence of buses, mopeds, medium vehicles, and trucks as traffic density increases. For example, from low to high traffic density, the relative amount of total counts for one or more buses increases from 6.7% to 24%. For mopeds and trucks the increase is 1.3 to 3.8% and 6.8% to 22%, respectively for low to high traffic density (see also Fig. S9).

Cycle paths that are located on the street also result in a 32% higher PNC than the ambient average, while cycle paths removed from the street and located on a shared pedestrian-cyclist sidewalk reduce PNC by 11% relative to the ambient average. The proximity to traffic and the overall amount and type of vehicular traffic have a significant influence on PNCs. The magnitude of influence of these types of factors should be considered when planning infrastructure, but could also be used by individuals to plan transit routes to minimize exposure to PM.

To compare with this work, a study that evaluated exposure to particles in 11 cities in the Netherlands cited an overall mean of 24,329 particles cm^{-3} for their bicycle measurements, with large variations observed within and between cities and sampling days (particle number concentration measured with a TSI CPC) (Boogaard et al., 2009). The mean value in this study was 7680 particles cm^{-3} (standard deviation: 3260 to 18,070 particles cm^{-3}) (PNC measured with a DiSCmini). Average bicycle trip PNCs were 22,660 particles cm^{-3} for a study conducted in Basel, Switzerland also using a DiSCmini (Ragetti et al., 2013). A study in Christchurch, NZ reported median PNC of 31,414 particles cm^{-3} and 16,641 particles cm^{-3} for on road and off road cyclists, respectively, using a TSI CPC (Kingham et al., 2013). The median PNC of this study was substantially lower at 7020 particles cm^{-3} . These comparisons show that overall mean and median particle number concentrations in Berlin were somewhat lower than those recorded in other cities. Our results indicate that even for cities with relatively moderate particulate levels, the specific location and proximity to vehicles makes a measurable difference to exposure rates.

A limited number of studies evaluated the effect of different variables, such as environment or cycling location and the effect these had on particle number concentrations. In a study evaluating the difference of bicycle commuting routes in Basel, Switzerland – one along main roads, and the other away from main roads – results showed that daily UFP exposure could be reduced by half if main roads were avoided (Ragetti et al., 2013). While we do not evaluate daily exposure in this study, we can compare the relative percent difference in PNC residuals between cycling on main roads (ST_MAINRD) to cycling on residential streets (ST_RESID), for which we found a decrease of ca. 36%. While this is similar to Ragetti et al. (2013), these values reflect only differences in PNC during cycling, whereas their measurements spanned 24 h, of which the commuting routes were only a small contribution in terms of time, although likely a large contribution to overall exposure.

The 11 cities study from the Netherlands evaluated the percent change in particle number concentration for a variety of predictor

variables during their occurrence relative to the total observation period. This is most similar to the approach taken here among existing studies, and one of the only other studies to quantitatively assess the effect of multiple variables influencing PNC. They observed an 11% increase for cycling on an on-road bicycle lane (lane separated from road vehicles by line marking only), and a smaller increase of 8% for cycling on a bike path for cyclists that is parallel but unattached to the road (Boogaard et al., 2009). In our study, we observed an increase of 32% in the cycling location variables for a cycle path on the street (CL_STRPATH), and a decrease of –11% for a cycle path on sidewalk (CL_SIDEWALK) relative to the average ambient level. The larger increase relative to the estimated background in our results may reflect the difference in methods, but could also be attributed to the amount of traffic on those routes where the cycle path is located on the street. That we see a decrease for cycle paths located on the sidewalk, whereby Boogaard et al. (2009) see a similar magnitude increase could be attributed to a number of factors, including differences in infrastructure between the countries/cities in terms of the distance of the sidewalk from the road, a variation in the type of road and/or traffic density for which cycle paths are placed on the sidewalk, etc. Among other comparable variables that they investigated were the increase resulting from passing mopeds (58%) and while waiting for traffic lights (10%) (Boogaard et al., 2009). Here we observed an increase of 38% owing to presence of mopeds (MOPED_1) and a 35% increase for waiting at traffic lights relative to the ambient average (ENV_TRAFLIGHT). The general consistency in these results from different cities indicates that such factors are likely to have similar effects in other cities as well.

These results, summarized in Table 2, highlight the substantial differences in particle number concentrations that exist over small scales, both temporally and spatially. These variations have significant implications for cyclists, but also for other urban citizens and both their activity and their transport choices. From these results we can conclude that infrastructure choices – for example, in terms of where and how cycling lanes are built – as well as their proximity to traffic (emissions sources) will have a large effect on the particle number concentrations that cyclists are exposed to. Furthermore, the traffic mix in the city and the proximity of these vehicles to cyclists can exacerbate short-term exposure to higher concentrations. The extensive number of variables investigated and quantified provides a wealth of information for possible consideration in urban planning. Results, while collected by bicycle, have direct implications for city dwellers' exposure rates due to local sources for a full range of activities, including walking and a range of recreational activities. While the general conclusions from this study are likely applicable in other cities given the general consistency of limited comparisons available, with variations in magnitude owing to local conditions, such as built environment and local geography expected, further such studies would allow for a better understanding of the full transferability of the results among cities. In terms of the built environment, the differences in the width and height of street canyons from the surrounding buildings would affect dispersion and thereby the PNC measured. This was not a variable that was classified in this study but could add additional information to future work. The lack of nearby stationary measurement site(s) with equivalent measurements was a limitation that was overcome by the development of an alternative background assessment method. However, future studies would be served by having such measurements available. The rapid changes in ultra-fine particle concentrations should be taken into account if a stationary site is used. The approach of non-prescribed bicycling routes and the resulting broad variability in locations for the different days measured results in a less consistent dataset. The local background correction also limits any comparisons across regions of the city. This research is therefore a more exploratory study. Furthermore, the evaluation of video data is time consuming and may include some inaccurate classifications or subjectivity as it is carried out by individuals. This will likely not influence the outcome with a

sufficiently large data set and clear classification guidelines, as was the aim here. Other options to address these issues would be automated video evaluation or possible crowd-sourcing. Finally, similar measurements carried out during other seasons would add value to such a dataset.

5. Conclusions

This exploratory study quantifies the effect of nearby pollution sources in urban areas, with a particular focus on quantifying the effects of a wide range of variables – cycling location, environment, presence and density traffic and vehicle types – on the particle number concentrations bicyclists are exposed to. These results have implications, not just for cyclists, but also for pedestrians and urban infrastructure more generally because the differences in local pollution levels are found to be quite large. The results show that buses (BUS_1) and trucks (TRUCK_1) have a larger contribution to PNC (39% and 41% increase over the ambient average, respectively) than cars (CAR_1, 12% increase and CAR_2, 32% increase over the ambient average). The overall traffic density also makes a difference to PNC the impact of low to high traffic density ranging from a 16% to 46% increase relative to the ambient average PNC, respectively. Furthermore, increased PNC for crossing intersections (ENV_INTERSEC, +38%), and being on cycle paths that are part of the main roadway (CL_STRPATH, +32%) also have a quantifiable impact on particulate matter exposure. These results are policy relevant and have implications for how we plan and build cycling infrastructure. For example, in order to minimize the exposure of cyclists to ambient particulate matter, cycle routes would ideally be constructed to avoid main roads, especially those with high traffic densities and/or routes with significant usage by larger trucks and buses. Shared bus and bicycle lanes are likely not an ideal option given these findings. Furthermore, where possible, the routes should pass through green spaces that further remove cyclists from traffic emissions and contribute air pollution filtering effects (e.g., Abd El Aziz et al., 2015; Selmi et al., 2016), as shown in this study. Current cycle infrastructure is often along main thoroughfares, especially in dense urban areas (e.g., Boogaard et al., 2009). The often conflicting priorities of creating the most direct routes and those that might reduce exposure by avoiding main thoroughfares will be a challenge to consider. The quantification of the impact of local traffic on bicyclists can be extrapolated to pedestrians and more generally, to outdoor activity within the urban area. A variety of factors will need to be considered in such urban planning to be able to create infrastructure that fits the needs of the different groups in a city while also, ideally, taking into account the implications for exposure to air pollution. We hope that by providing information on a number of the factors influencing air pollution PNC here, that such factors can be considered when evaluating the potential effectiveness of policies under consideration. The identification of the magnitude of impact of local traffic and road proximity can allow for evaluation of options for cities that includes the impact on human health for different development options.

Acknowledgements

This work was hosted by IASS Potsdam, with financial support provided by the Federal Ministry of Education and Research (BMBF), Germany and the Ministry for Science, Research and Culture of the State of Brandenburg (MWFK), Germany. We would like to thank our colleagues at the IASS that participated in the bicycle measurements, Aditi Rosegger for video evaluation, Mark Lawrence for his support of the campaign and discussions on the data, as well as colleagues at the Research Center Jülich, German Environment Agency, Finnish Meteorological Institute, Senate Department for the Environment, Transport and Climate Protection in Berlin, Karlsruhe Institute of Technology, and University of Wisconsin-Madison for their participation in and support throughout the campaign.

Appendix A. Supplementary information

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2019.06.309>.

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