# **Environmental Monitoring and Assessment**

# Cyclists' exposure to air pollution: In situ evaluation with a cargo bike platform -- Manuscript Draft--

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Abstract:	Cyclists' exposure to air pollutants near roadways has been associated with numerous health effects. While, the adverse health effects concerning aerosols have traditionally been assessed with data of particle mass concentrations, it appears that the number concentration is also another important indicator of toxicity. Thus, to holistically evaluate one's exposure to aerosol particles, assessments should be based on mass concentrations and number concentrations. In order to assess individual cyclists' exposure as they move through space and time, spatiotemporal high-resolution approaches are needed. Therefore, a mobile, fast-response monitoring platform was developed that uses a cargo bicycle as a base. Data of particle mass concentrations (PM 1, PM 2.5, PM 10) and particle number concentrations (PN 10) were collected along two different routes, one characterized by high-intensity vehicle traffic and one by low-intensity vehicle traffic. While high spatiotemporal heterogeneity was observed for all measured quantities, the PN 10 concentrations fluctuated the most. High concentrations of PN 10 could be clearly associated with vehicle traffic. For PM 2.5, this relation was less pronounced. Mean particle concentrations of all measures were significantly higher along the high-traffic route. Comparing route exposures, the inhalation of PM 2.5 was similar between both routes, whereas along the high-traffic route, cyclists were exposed to twice the particle number. We conclude that the cargo bike, featuring high-frequency mobile measurements, was useful to characterize the spatial distribution of mass concentrations and number concentrations across an urban environment. Overall, our results suggest that the choice of route is a key factor in reducing cyclists' exposure to air pollution.
Response to Reviewers:	Reviewers' Comments to the Authors:  1.In my opinion, the results and discussion chapter should be divided into separate subsections: results and discussion.  Author response: As suggested by the reviewer, we have split the results and discussion section into separate subsections.

2. Conclusions should be shortened and the fragments moved to discussion. Author response: Thank you for the suggestion. Some paragraphs were moved to discussion. We keep in this section only the main central facts of our study, that were previously argumentized in the discussion section.

3.In Fig. 1 small and heavy traffic routes should be marked.
Author response: Thank you for the suggestion. Primary and secondary routes were indicated in the map.



To the Associate Editor of Journal Environmental Monitoring and Assessment

Ref: EMAS-D-19-04262

Dear Dr. Gregory J. White

We appreciate the time and effort that you and the reviewer dedicated to providing feedback on our manuscript and are grateful for the valuable comments on our paper. We have incorporated all the corrections suggested by the reviewer. These changes are highlighted within the manuscript. Please see below, a point-by-point response to the reviewers' comments.

## **Reviewers' Comments to the Authors:**

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Sincerely yours,

Hebe Carreras, PhD. Corresponding author

1 Cyclists' exposure to air pollution: *In situ* evaluation with a cargo bike platform 2 3 Hebe Carreras a\*, Laura Ehrnsperger b, Otto Klemm b, Bastian Paas b 4 5 <sup>a</sup>Instituto Multidisciplinario de Biología Vegetal, CONICET and Chemistry Department, 6 FCEFyN, Universidad Nacional de Córdoba, Av. Velez Sarsfield, 1611, X5016 GCA Córdoba, 7 Argentina. 8 9 <sup>b</sup>University of Münster, Climatology Research Group, Heisenbergstr. 2, 48149 Münster, Germany 10 11 \*Corresponding author. E-mail address: hebe.carreras@unc.edu.ar (H. Carreras). Tel.: +54 351 12 5353800 int. 29771. 13

#### **Abstract**

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Cyclists' exposure to air pollutants near roadways has been associated with numerous health effects. While, the adverse health effects concerning aerosols have traditionally been assessed with data of particle mass concentrations, it appears that the number concentration is also another important indicator of toxicity. Thus, to holistically evaluate one's exposure to aerosol particles, assessments should be based on mass concentrations and number concentrations. In order to assess individual cyclists' exposure as they move through space and time, spatiotemporal highresolution approaches are needed. Therefore, a mobile, fast-response monitoring platform was developed that uses a cargo bicycle as a base. Data of particle mass concentrations (PM<sub>1</sub>, PM<sub>2.5</sub>,  $PM_{10}$ ) and particle number concentrations (PN<sub>10</sub>) were collected along two different routes, one characterized by high-intensity vehicle traffic and one by low-intensity vehicle traffic. While high spatiotemporal heterogeneity was observed for all measured quantities, the PN<sub>10</sub> concentrations fluctuated the most. High concentrations of PN<sub>10</sub> could be clearly associated with vehicle traffic. For PM<sub>2.5</sub>, this relation was less pronounced. Mean particle concentrations of all measures were significantly higher along the high-traffic route. Comparing route exposures, the inhalation of PM<sub>2.5</sub> was similar between both routes, whereas along the high-traffic route, cyclists were exposed to twice the particle number. We conclude that the cargo bike, featuring high-frequency mobile measurements, was useful to characterize the spatial distribution of mass concentrations and number concentrations across an urban environment. Overall, our results suggest that the choice of route is a key factor in reducing cyclists' exposure to air pollution.

#### 34 Keywords

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Cargo bike, exposure assessment, particle number concentration, particle mass concentration

#### Introduction

37 Bicycling is – besides walking – widely accepted as the most environmentally friendly means of 38 transport since it does not contribute to air or noise pollution nor to any extra emission of carbon 39 dioxide. Cycling also provides many health benefits such as improving cardiovascular health, 40 muscular strength, and control of blood sugar (Bauman and Rissel, 2009, Kelly et al., 2014, Oja 41 et al., 2011). However, cycling has also been related to many negative health issues due to 42 cyclists' exposure to high levels of air pollutants and noise (Boogaard et al., 2008) when traveling 43 on roads shared with motor vehicles or on routes adjacent to or near main roads (Badland and 44 Duncan, 2009; De Hartog et al., 2010; Kaur et al., 2007). The typical exposure of cyclists is 45 relatively short, but the dose is high due to their higher breathing activity compared to those of 46 car or bus passengers (Boogaard et al., 2008, Brand et al., 2019). Consequently, cyclists' exposure 47 to both gaseous and particulate air pollutants near roadways has been associated with numerous 48 health effects including asthma exacerbation, other respiratory illnesses, and excess risk of 49 mortality from cardiopulmonary disease and stroke (Apparicio et al., 2016; Gordian et al., 2006). 50 Traditionally, the adverse health effects of particulate matter (PM) have been associated with 51 PM<sub>10</sub> or PM<sub>2.5</sub> particles, i.e., the mass concentrations of PM with aerodynamic diameters below 52 10 µm and 2.5 µm, respectively. Yet, the specific health effects of ultrafine particles with an 53 aerodynamic diameter  $\leq 0.1 \, \mu m$  (UFP), which do not contribute much to the PM<sub>2.5</sub> or PM<sub>10</sub> mass 54 concentrations, are still under discussion. For example, in 2013 the Health Effects Institute 55 concluded that the state of science was inconclusive regarding the toxicity of UFPs due to 56 inconsistencies and limitations in the findings from both short-term and long-term studies (EPA, 57 2015). While some studies state that the total particle number concentration in air, which is a good 58 indicator of UFP, remains a poor indicator of toxicity markers (Xue et al., 2019), others suggest 59 that UFPs cause a greater lung inflammatory response than is caused by equal mass concentrations 60 of larger particles (Frampton, 2016). Similarly, Brown et al. (2001) and Oberdorster (2001) 61 reported that UFP exhibited greater pro-inflammation activity per mass than the same mass of 62 larger particles. Overall, it appears that the mass concentration of UFP exhibits fewer toxic effects 63 than the number concentration of UFP (Penttinen et al., 2001; von Klot et al., 2002; Ferin et al., 64 1992, Hosiokangas et al., 1995). Consequently, we hold the view that any evaluation of cyclists' 65 exposure to aerosol particles should be based both on mass concentration (PM<sub>10</sub>, PM<sub>2.5</sub>) and 66 number concentration (PNC) data.

Within cities, motor traffic has been identified as a major contributor to the near-road aerosol strain (Karagulian et al., 2015; Manousakas et al., 2017) due to both exhaust and non-exhaust sources (Ketzel et al., 2007; Amato et al., 2014). Raised levels of particle concentrations mostly occur within street canyons (Wurzler et al., 2016). Further, there are diffuse sources of aerosol particles such as resuspension from dry, grassy and unpaved footpaths containing loose and dry top coating material (Birmili, 2013; Paas et al., 2016). Further, Birmili et al., 2013 identified cooking activities and smoking in outdoor restaurant seating areas to be important sources of PM. The combination of various particle sources and the influence of meteorological conditions on the transport of aerosols in the urban boundary layer poses a challenge to determining aerosol concentration patterns (Venkatachari et al., 2006).

An increasing number of studies have used simulation approaches (Baxter et al., 2010) or land use regression and related models (Gryparis et al., 2007) to estimate the spatiotemporal distribution of air pollutants for use in epidemiological studies. While these studies have provided insight about exposure patterns and health effects, they lack sufficient spatial resolution to capture the adverse exposure dose on the micro scale. Further, some of the studies provided significant misclassifications of exposure, which tends to obscure true correlations between air pollutant concentrations and potential health effects (Baxter et al., 2010; Sarnat et al., 2010).

When it comes to individual exposure, the situation is even more complex for subjects moving through space and time (Kousa et al. 2002). With each breath of air, another air mass, with potentially much different concentrations from the earlier or later breath, respectively, is inhaled. In the Netherlands, a tool has been established to assess local cycling conditions, and this tool also includes the evaluation of the current air quality (Boogaard et al., 2008). Commuters can use the spatial information on air pollution to avoid high exposure by choosing less-polluted routes. In addition, urban planners and policymakers can make use of the detailed spatial information to identify local hotspots and to take measures against local air pollution.

However, to evaluate local air pollution, such tools primarily use stationary air quality monitoring networks that only report 30-minute or 60-minute averages. These networks measure - following legal requirements - particle mass concentrations (PMC, either PM<sub>10</sub> or PM<sub>2.5</sub>) but not PNC. Further, such stations cannot account for the high variability of air pollutant concentrations in urban environments. For these reasons, no reliable assessment of an individual cyclist's exposure can be made from the respective data. In fact, the exposure to air pollutants in traffic, which is found to be generally enhanced, is, in many cases, only poorly correlated to simultaneous measurements at stationary monitoring sites (Kaur et al., 2007). Therefore, there is a clear need for measurements of air pollutants to be taken at much better spatial and temporal resolutions of 1 m and 1 s, respectively, in order to evaluate cyclists' exposure.

During the last decade, mobile monitoring platforms enabling personalized and high-resolution monitoring have become increasingly popular (Broich et al., 2012; Birmili et al., 2013; Van den Bossche et al., 2015; Dewulf et al., 2015, Gerike et al., 2016). Especially for atmospheric pollutants that experience large spatiotemporal variation in urban environments, mobile monitoring provides valuable insights into the spatial distribution of air pollution that cyclists and pedestrians are exposed to in everyday life. However, to the best of our knowledge, no experimental study has simultaneously characterized the spatiotemporal patterns of both particle mass concentration and particle number concentration in an urban environment. Thus, we used a mobile cargo bike platform to a) assess the potential of real-time PMC and PNC monitoring to measure the exposure of cyclists who commute daily in an urban environment, b) to provide insight into the spatiotemporal variability of fine and ultrafine particle concentrations, and c) to quantify the potential human health risk due to exposure to fine particles depending on the traffic density of the route chosen.

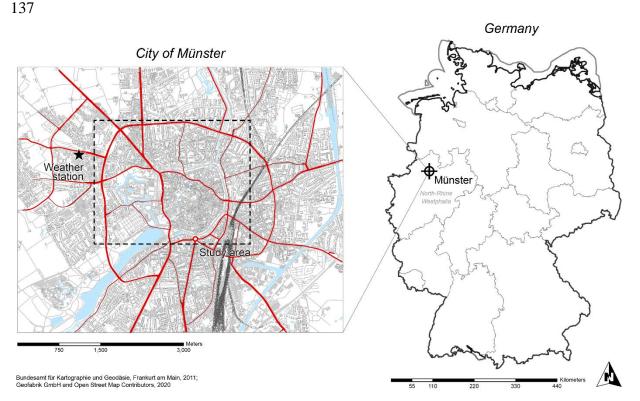
#### Methods

116 Site description

This study was carried out in Münster, NW Germany (52.0 °N, 7.6 °E), a city with approximately 300,000 inhabitants (Fig. 1). It is characterized by flat terrain and hosts some small- to medium-sized industries. The city is well documented in terms of traffic flow, and there is a good understanding of the underlying processes driving air pollution (Gietl and Klemm, 2009). The regional impact of air pollution originates from long-range transport, mainly from the Ruhr industrial area located 60 km to the SW since the main wind direction is southwest, as determined over the period from 1989 – 2018 (DWD, 2019), and from the intense agricultural activity of the surrounding area (Gietl et al. 2008; Gietl and Klemm 2009). Local sources are traffic emissions, residential heating, and to a lesser degree industry and power plant emissions (Deventer et al., 2015).

Since almost 40 % of daily routes are made by bike in Münster, we selected two routes with different traffic influence, crossing the city from east to west. The high-traffic route (2.45 km) goes along a main thoroughfare that links the central train station to the downtown area and eventually leads to the university campus, while the low-traffic route (2.99 km) goes through a green corridor that surrounds the downtown area, and which may only be used by bicyclists and pedestrians. The two courses were probed in the morning of July 17<sup>th</sup>, 2019, between 09:50 and 10:35 local time (UTC + 2). Average synoptic conditions during data collection were a northwest

(290°) wind direction and a wind speed of 1.6 m s<sup>-1</sup>, as observed at the weather station at the university campus at 34 m above ground, 1800 m beeline from the downtown area (Fig. 1).



**Fig. 1** Study area within the city center (dashed bordered rectangle) of Münster, Germany and location of the weather station (star). Main traffic arterials are marked with red lines (bold: primary roads, narrow: secondary roads).

#### Data collection and processing

Data were collected with a cargo bike equipped with a three-dimensional sonic anemometer, type R3-50 (Gill Instruments Ltd., Lymington, Hampshire, UK). The anemometer was installed vertically on the front end of the cargo bike with the center of the sensor head being at a height of 1.47 m above ground (Fig. 2). The North arrow of the anemometer was aligned with the direction of driving to enable correction of wind direction in post-processing. The aerosol particle measurement technology included an optical particle spectrometer (OPS), type 3330, and a condensation particle counter (CPC), type 3025A (both sensors manufactured by TSI Inc., Minnesota, USA). The OPS sampled the PMC in 16 size classes (0.3 to 10 μm diameters), while the CPC sampled the total PNC of particles between 3 nm and 10 μm diameters (PN<sub>10</sub>). The upper detection limit of the CPC was 53,000 p cm<sup>-3</sup>, meaning that a concentration higher than 53,000 particles per cm<sup>3</sup> could not be detected. During post-processing, the respective PMCs of individual size classes were summed up to the most common measures of PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1</sub>. The air samples were taken at the front end of the cargo bike at a height of 1.1 m above ground

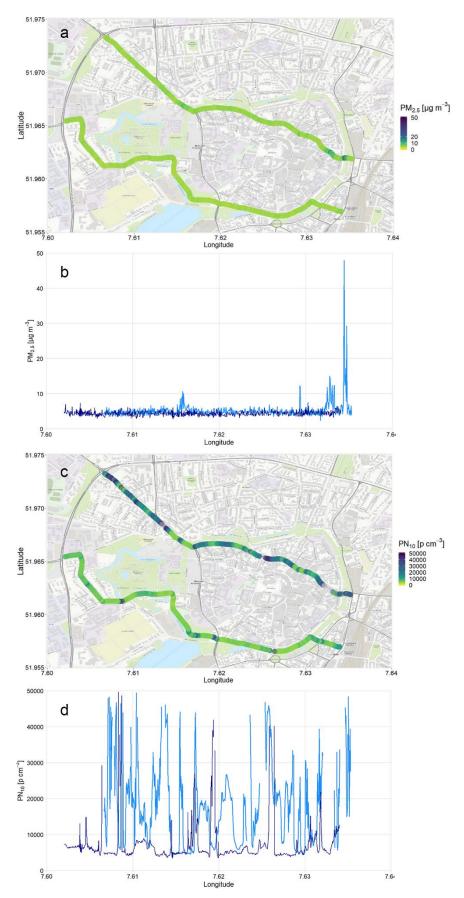
155 and led through individual hoses of 0.96 m (internal diameter: 5 mm) and 1.28 m (internal 156 diameter: 10 mm) lengths to the OPS and CPC, respectively. To account for the position and 157 ground speed as well as for the orientation of the cargo bike, a GPS device, type 19x HVS (Garmin 158 Ltd., Kansas, USA) and a digital compass module, type HMR3000 (Honeywell International Inc., 159 North Carolina, USA) were used ( 160 Fig. 2). All sensors were operated with 1 Hz data acquisition frequency, centrally controlled by a 161 data logger, type CR3000 (Campbell Scientific Inc., Utah, USA). 162 Processing of raw data was made with R version 3.5.0 (R Core Team, 2018). The R packages 163 ggplot2 3.0.0 (Wickham, 2016) and OpenStreetMap 0.3.4 (Fellows, 2019) were used for the 164 cartographic display of the data.



167 **Fig. 2** Cargo bike with a) ultra-sonic anemometer, b) sample inlet for CPC, c) sample inlet for OPS, d) GPS antenna, e) digital compass, and f) aluminum box containing the instruments and power supply.

#### 170 **Results**

171 Spatiotemporal variability of aerosol particle concentrations 172 The concentrations of PM<sub>2.5</sub> and PN<sub>10</sub> as measured along the two cargo bike routes are shown as 173 cartographic displays and as longitude series in Fig. 3. The two routes that lead from east to west 174 through the city of Münster are primarily different in their influence of motor vehicle traffic. The 175 northern route experienced high vehicle traffic (HT), whereas the southern route was influenced 176 by low traffic intensities (LT). 177 The second-to-second differences can be as high as 28.5 µg m<sup>-3</sup> and 43,500 p cm<sup>-3</sup> for PM<sub>2.5</sub> and 178 PN<sub>10</sub>, respectively. Comparing the PM<sub>2.5</sub> concentration data from the two routes side by side, it 179 becomes apparent that the median values are quite similar for both routes (LT: 4.3 µg m<sup>-3</sup>, HT: 180 4.7 µg m<sup>-3</sup>). Nevertheless, the HT route exhibited considerably more variation. The mass concentration range was between 2.8  $\mu g$  m<sup>-3</sup> and 7.3  $\mu g$  m<sup>-3</sup> in the LT route and between 2.4  $\mu g$ 181 182 m<sup>-3</sup> and 48 µg m<sup>-3</sup> in the HT route. The PM<sub>2.5</sub> concentrations did not vary largely along the LT 183 route. However, five hotspots were identified along the HT route; Apart from these few hotspots, 184 the impact of traffic on the observed PM<sub>2.5</sub> concentrations was detectable yet small. Road traffic 185 is only a minor source of PM<sub>2.5</sub> compared to long-range transport of particles.



**Fig. 3** PM<sub>2.5</sub> concentrations ( $\mu$ g m<sup>-3</sup>) as a) a color-coded cartographic display and as b) a longitude series; PN<sub>10</sub> concentrations (p cm<sup>-3</sup>) as c) a color-coded cartographic display and as d) a longitude series. Data were collected at 1 Hz along two routes characterized by high traffic influence

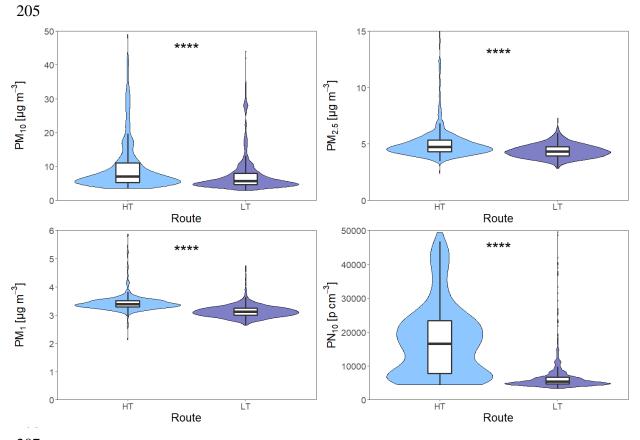
(northern route, light blue line) and by low traffic influence (southern route, dark blue line) in the city of Münster, Germany.

More pronounced differences between the two routes were observed for  $PN_{10}$  compared to  $PM_{2.5}$ . For the LT route, the particle number concentration ranged between 3,400 p cm<sup>-3</sup> and the value for the maximum detectable concentration, 53,000 p cm<sup>-3</sup>, (median: 5,400 p cm<sup>-3</sup>), and for the HT route it ranged between 4,400 p cm<sup>-3</sup> and 53,000 p cm<sup>-3</sup> (median: 17,000 cm<sup>-3</sup>).

The spatial analysis of both the HT and LT routes concerning  $PN_{10}$  unveils that the spatial variations of  $PN_{10}$  concentrations were very high (Fig 3 c). Furthermore, elevated  $PN_{10}$  concentrations could almost always be associated with areas such as street intersections and sectors around traffic lights and, thus, with motorized vehicle traffic.

A cyclist's exposure dose on individual routes

On average, cyclists were exposed to higher PMC and PNC when biking along the HT route as compared to the LT route. The differences were much larger when comparing  $PN_{10}$  particle number concentrations than  $PM_{10}$  mass concentrations, and they were also larger when comparing larger rather than smaller size fractions of particles (Fig. 4).



**Fig. 4** Box-Whisker and violin plots for mass concentrations of  $PM_{10}$ ,  $PM_{2.5}$  and  $PM_1$  and  $PN_{10}$  for the high-traffic (HT) and low-traffic (LT) routes. The boxes represent the interquartile range (IQR,  $25^{th}$  to  $75^{th}$  percentile); whiskers represent the  $10^{th}$  and  $90^{th}$  percentiles. The colored areas show the kernel probability density of each parameter. Outliers are not shown for clarity. The stars in each plot represent the significance level (Wilcoxon Rank Test, p-value < 0.0001 = \*\*\*\*).

The PMCs of PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1</sub> showed larger variations for the HT route than for the LT route; the variation coefficients (CV) were larger by factors of 1.34, 3.32 and 2.33, respectively. The spatial analysis revealed that variations of PM<sub>2.5</sub> are weakly associated with intersections between collector or arterial roads (Fig. 3 a). No such difference of CVs between the HT and LT routes was observed for the particle number concentrations (CV HT route: 63.28; CV LT route: 75.04). Overall, the high variability of PN<sub>10</sub> as compared to PMC is associated with traffic emissions. The more pronounced variability of PN<sub>10</sub> along the LT route is caused by the generally lower baseline of PN<sub>10</sub> concentrations in combination with the crossing of a few street intersections, where the maximum PN<sub>10</sub> concentrations were as high as the maximum concentrations detected along the HT route (Fig. 3 c, d).

Our results show much higher concentrations of PM<sub>10</sub> and PN<sub>10</sub> along the HT route than along the LT route, while levels of PM<sub>2.5</sub> and PM<sub>1</sub> were also higher for the HT route, but to a lesser degree. In order to arrive at a quantitative estimation of cyclist's exposition while riding along different route types, we quantify the route exposure (RE) to PM<sub>2.5</sub> as follows:

229 
$$RE(\mu g) = concentration\left(\frac{\mu g}{m^3}\right) \times inhalation \, rate\left(\frac{m^3}{s}\right) \times time \, lapse(s)$$

and to the particle number p as

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$$RE(p) = concentration\left(\frac{p}{m^3}\right) \times inhalation\ rate\left(\frac{m^3}{s}\right) \times time\ lapse(s)$$

We assumed an average volumetric inhalation rate of  $0.065~\text{m}^3~\text{min}^{-1}$  for cyclists, based on data from the Exposure Factor Handbook (Environmental Protection Agency, 2011). The overall exposure of each route was calculated with the mean mass of particles <  $2.5~\mu m$  inhaled as well as the total number of particles <  $10~\mu m$  inhaled as metrics to estimate the personal exposure to  $PM_{2.5}$  and  $PN_{10}$ , respectively. We found that cyclists using the HT route inhaled 4.22  $\mu g$  of particles <  $2.5~\mu m$ , while when using the LT route, which lasted longer, they inhaled 4.86  $\mu g$  of particles <  $2.5~\mu m$ . Thus, even though the LT route had lower  $PM_{2.5}$  concentrations, this was outweighed by its longer distance and, thus, its longer travel time (1,010 s vs 741 s). Note that a cyclist traveling on the HT route also needs to breathe air when not biking (and this air potentially contains  $PM_{2.5}$  as well), such that his total exposure during the entire time period of the LT route will be somewhat larger than his calculated HT exposure. For both routes, then, the exposure to

- PM<sub>2.5</sub> is quite similar. However, when assessing the cyclists' exposure to particle number, we found that cyclists using the HT route were exposed to twice the particle number as compared to those using the LT route  $(14,700 \times 10^6 \text{ vs. } 7,500 \times 10^6 \text{ particles})$ . Thus, as this particle fraction and metric are more relevant to toxicological effects, the toxicological risk to a cyclist on the HT route is at least twice as high as the toxicological risk to a cyclist on the LT route.
  - Discussion

- The overall spatiotemporal variabilities of the measured  $PM_{2.5}$  and  $PN_{10}$  concentrations at both sampling routes are very large. Boogaard et al. (2008) also found very high variability for one-
- 251 minute averages of PM<sub>2.5</sub> and PNC within eleven Dutch cities. They were able to demonstrate that
- 252 peaks of PNC occurred more often and more suddenly for cyclists than for car drivers with a
- 253 typical peak length of less than 10 s (Boogaard et al., 2008).
  - However, the PM<sub>2.5</sub> concentrations measured at the HT and LT routes are much lower than those measured by Ham et al. (2017), who compared daily commutes using bike trails or arterial trails in Sacramento, CA ( $6 \pm 1.5 \,\mu g \, m^{-3}$  and  $13.5 \pm 7 \,\mu g \, m^{-3}$ , respectively). Boogaard et al. (2008) also found higher mean PM<sub>2.5</sub> concentrations (44.5  $\,\mu g \, m^{-3}$  for cyclists), albeit the variability of concentrations in their one-minute data set is similar to the range we found in our Münster one-second dataset (5 to 112  $\,\mu g \, m^{-3}$ ). The PM<sub>2.5</sub> hotspots identified along the LT route can be spatially associated with areas likely to be affected by high traffic intensities at intersections or traffic lights. These areas typically experience stop-and-go vehicle traffic and, therefore, particles are emitted from brake and tire abrasion, which contributes to PM<sub>2.5</sub> mass concentrations (Ketzel et al., 2007; Amato et al., 2014). Further, as is known from other studies, the particle mass concentration in the size range of PM<sub>2.5</sub> is heavily influenced by accumulation mode particles that originate from long-range transport (Gidhagen et al., 2004; Ketzel and Berkowicz, 2005, Gietl et al. 2008; Gietl and Klemm 2009). Our findings confirm that compared to the long-range transport sources, local vehicle traffic sources are evident but less important in the case of PM<sub>2.5</sub>.
  - Regarding differences in PN<sub>10</sub> concentrations between the sampling routes, our findings agree well with the mean UFP concentrations as measured by Ham et al. (2017) for bike trails and arterial trails in Sacramento, CA (10,000 p cm<sup>-3</sup> to 24,000 p cm<sup>-3</sup>), with the results from Vinzents et al. (2005) for data collected in central Copenhagen (geometric mean: 32,400 p cm<sup>-3</sup>), and with a study from Thai et al., 2008 that was conducted in Vancouver (UFP concentration range: 18,830 p cm<sup>-3</sup> to 57,692 p cm<sup>-3</sup>). Our resulting ratio of three to one when comparing the PN<sub>10</sub> medians of the HT and LT routes agrees well with findings from Ham et al. (2017). The authors of the Sacramento study attributed the differences of mean UFP concentration to the proximity of

the bike route to the motorized traffic and to differences in vehicle activity between these two routes. Other authors also published similar ratios despite having different study areas (Jarjour et al., 2013; Zuurbier et al., 2010).

279 Compared to the spatial concentration variability of PM<sub>2.5</sub>, the measured concentrations of PN<sub>10</sub> 280 showed a largely different picture. Particle number concentrations are heavily influenced by short-281 lived particles within the nucleation mode that originate both from primary emissions and from 282 secondary formation from precursor gases from combustion processes (Ning and Sioutas, 2010; 283 Quiros et al., 2013). Kingham et al. (2013) reported median  $PN_{10}$  of 31,414 particles cm<sup>-3</sup> and 284 16,641 particles cm<sup>-3</sup> for on-road and off-road cyclists, respectively, in Christchurch, New 285 Zealand, showing that the overall mean PN<sub>10</sub> concentrations detected in this study (18,026 and 286 6,700 particles cm<sup>-3</sup>) are rather low in comparison with other cities. Boogaard et al. (2008) also 287 showed that factors like the type of passing vehicles, the time spent waiting at traffic lights and 288 intersections, the type of intersection, and the distance of bike lanes to motorized traffic routes all 289 contributed to the variability of particle number concentration in eleven Dutch cities. Similarly, 290 Pattinson et al. (2017) found 40 to 50 % lower ultrafine particle concentrations on a path that was 291 19 m away from the road than on the main road itself in Christchurch; and Brand et al. (2019) 292 found a rise in black carbon average concentration when the cycle lane inside Hyde Park 293 approaches to the edge of the park, which was attributed to the proximity of traffic emission 294 sources. Our PN<sub>10</sub> results confirm that local vehicle traffic sources play a major role for local PN<sub>10</sub> 295 concentrations.

The analysis of cyclist's exposure dose on individual routes, shows much higher concentrations of  $PM_{10}$  and  $PN_{10}$  along the HT route than along the LT route. Similarly, MacNaughton et al. (2014) found that bike paths have lower concentrations of traffic-related pollutants than bike lanes along heavy traffic routes. Ham et al. (2017) found that average  $PM_{2.5}$  concentrations were lowest for trips taken on a dedicated bike trail. We believe that the health risk for cyclists is better explained by exposure to UFP rather than to  $PM_{10}$ , because a high number concentration of UFP is of higher toxicological relevance than a high  $PM_{10}$  mass concentration (Penttinen et al., 2001; von Klot et al., 2002).

The route exposure metrics reveals that cyclist exposure to PM<sub>2.5</sub> is quite similar between routes.

However, since cyclists' exposure to particle number at the HT doubled the one at the LT route,
the toxicological risk was twice larger at the former route. Similarly, Lonaty et al., (2016) stated
that the higher the volume of traffic, the greater a cyclist's exposure to traffic-related pollutants,
in particular UFP. Our results confirm that vehicle traffic intensities play a major role for local
UFP concentrations, while the influence of vehicle traffic sources on local PM<sub>2.5</sub> concentrations
is less pronounced.

is less pronounced.

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#### Conclusion

In this study, it was demonstrated that mobile direct measurements of particle mass concentration and particle number concentration are feasible at a high temporal resolution (1 Hz) without requiring any surrogate measures or assumptions. The monitoring platform mounted in the cargo bike was useful for characterizing the spatial distribution of PMC and PNC simultaneously across an urban environment, clearly showing the contrasting dynamics of PMC and PNC along various routes. Particle mass concentration and particle number concentration differed depending on the proximity to motorized traffic. The temporal as well as the spatial variability of PN<sub>10</sub> was very high, while the differences were less distinct for the PM<sub>2.5</sub> mass concentration.

Overall, our results suggest that the choice of route is a key factor for reducing cyclists' exposure to air pollution, although the duration and distance traveled can – for some metrics – offset the exposure reduction when using low-traffic routes. In our study for example, using the bike path through the city, which was well separated from the street, might result in a higher mass of  $PM_{2.5}$  particles inhaled, since this route was longer than the bike lane next to the street. However, the larger amount of UFP particles found on bike lanes adjacent to arterial streets indicates that even if cyclists reduce their exposure time, routes closer to traffic result in much higher levels of particle exposure.

We acknowledge some limitations in the present study. First, not every particle inhaled by a human will indeed reach the pulmonary alveoli; therefore, we might overestimate the particles' effects. On the other hand, we did not consider the chemical composition of particles by any means, which might have given more insight into specific toxic effects.

Our results stress the importance of studying the levels of UFP, which humans are exposed to. In our view, it is not sufficient to measure the particle mass concentrations (PMC). Legislation and policymakers should acknowledge the need to further improve environmental observation targets such as PNC in order to help mitigate humans' exposure, specifically cyclists' exposure, to air pollution.

We performed only a limited number of measurements. Many more trips with the instrumented cargo bike are required to arrive at statistically robust results concerning cyclists' exposure to air pollution in various cities, climates, traffic situations, commuting distances, and traveling speeds. In that sense, this is a feasibility study that proves the technical and scientific concept to be successful.

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#### **Conflict of interest**

349 The authors declare that they have no conflict of interest.

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