Exposure to ultrafine particles while walking or bicycling during COVID-19 closures: A repeated measures study in Copenhagen, Denmark

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HIGHLIGHTS

- Active mobility close to motorized traffic exposes people to high levels of UFP.
- COVID-19 closures reduced the need to commute and therefore local traffic counts.
- We measured UFP, walking and bicycling, during COVID-19 closures and re-opening.
- PNC levels decreased continuously during phased re-opening.
- PNC decreases mainly driven by meteorological factors (wind speed, temperature).

GRAPHICAL ABSTRACT
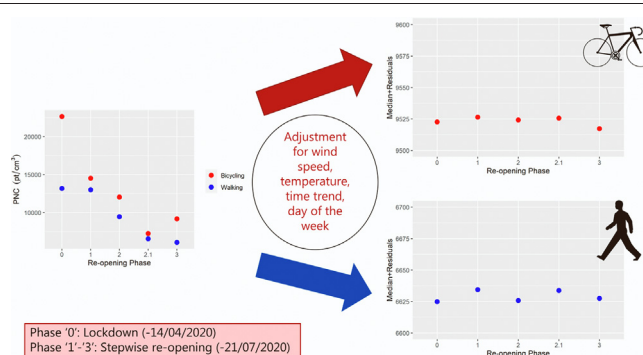
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ABSTRACT

Ultrafine particles (UFP; particulate matter <0.1 μm diameter) emitted from motorized traffic may be highly detrimental to health. Active mobility (walking, bicycling) is increasingly encouraged as a way to reduce traffic congestion and increase physical activity levels. However, it has raised concerns of increased exposure to UFP, due to increased breathing rates in traffic microenvironments, immediately close to their source. The recent Coronavirus Disease 2019 (COVID-19) societal closures reduced commuting needs, allowing a natural experiment to estimate contributions from motorized traffic to UFP exposure while walking or bicycling.

From late-March to mid-July 2020, UFP was repeatedly measured while walking or bicycling, capturing local COVID-19 closure ('Phase 0') and subsequent phased re-opening ('Phase 1', '2', '2.1' & '3'). A DiSCmini continuously measured particle number concentration (PNC) in the walker/bicyclist's breathing zone. PNC while walking or bicycling was compared across phased re-openings, and the effect of ambient temperature, wind speed and direction was determined using regression models.

Approximately 40 repeated 20-minute walking and bicycling laps were made over 4 months during societal re-opening phases related to the COVID-19 pandemic (late-March to mid-July 2020) in Copenhagen. Highest median PNC exposure of both walking (13,170 pt/cm^3, standard deviation (SD): 3560 pt/cm^3) and bicycling (21,477 pt/cm^3, SD: 8964) was seen during societal closures (Phase 0) and decreased to 5367 pt/cm^3 (SD: 2949) and 8714 pt/cm^3 (SD: 4309) in Phase 3 of re-opening. These reductions in PNC were mainly explained

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by meteorological conditions, with most of the deviation explained by wind speed (14–22%) and temperature (10–13%). Highest PNC was observed along major roads and intersections.

In conclusion, we observed decreases in UFP exposure while walking and bicycling during societal re-opening phases related to the COVID-19 pandemic, due largely to meteorological factors (e.g., wind speed and temperature) and seasonal variations in UFP levels.

1. Introduction

Air pollution is a major threat to public health worldwide, and the fourth leading contributor to disease burden worldwide according to the most recent Global Burden of Disease Study (Murray et al., 2020). Globally, ambient air pollution is responsible for about 4.5 million premature deaths every year (Murray et al., 2020), and 4200 in Denmark (Ellermann et al., 2020). Extensive research has been conducted especially on the health burden related to particulate matter (PM) of diameter <2.5 μm (PM$_{2.5}$) and nitrogen dioxide (NO$_2$) (European Environment Agency, 2020; Health Effects Institute, 2020). Due to a lack of regulation and measurements, evidence is lacking on the health effects related to ultrafine particles (UFP; diameter <0.1 μm). Toxicological evidence suggests that UFP are potentially more harmful to health than larger particles. This is largely due to their high surface-to-mass ratio and small size, allowing them to penetrate deeper into the lungs, epithelium, and translocate to the blood system and organs while carrying potential toxins via particle surface adsorption (Schraufnagel, 2020).

People experience peaks in daily exposure to air pollution, such as UFP, while they move through city streets, and in transport microenvironments close to motorized traffic. UFP levels in these microenvironments, for different modes of commuting and leisure time activities including walking or bicycling, may be orders of magnitude higher than those captured by annual average exposures at the home address or daily means of air pollution at city background monitors (de Nazelle et al., 2017). Furthermore, the choice of travel modes may significantly affect individual exposure to UFP (Knibbs et al., 2011). With the increasing interest in the promotion of active mobility through walking and bicycling, it is useful to quantify relative exposures to UFP in these travel mode microenvironments (Dons et al., 2015). Active mobility increases breathing rates while in close proximity to primary sources of UFP, consequently increasing concentrations of UFP taken into the body (Dons et al., 2017). This potential higher pollutant dose can have acute adverse effects such as reductions in lung function due to airway inflammation (Matt et al., 2016), discomfort for healthy individuals (Cole-Hunter et al., 2013) or worsened symptoms for asthmatic individuals (McCreanor et al., 2007). While physical activity such as that derived through active mobility should be encouraged, air pollution concentrations in this microenvironment should be reduced such as by reducing proximity to or volume of motorized traffic.

The recent Coronavirus Disease 2019 (COVID-19) pandemic provided a natural experiment (or counter-factual) for reduced traffic volumes through the imposition of societal closures in many cities around the world. These closures of workplaces and schools largely reduced the need to commute and therefore decreased motorized traffic-related emissions, including UFP (Hudda et al., 2020).

Copenhagen is a northern European city with a population of around 640,000 (Statistics Denmark, 2021). Active mobility, such as cycling, rates in Copenhagen are among the highest worldwide, with approximately 49% of commuting trips to work and school made by bicycle (City of Copenhagen, Technical and Environmental Administration, (TMF), Mobility, 2019). Our study, which is opportunistic in nature, aimed to assess how exposure to traffic-related UFP among active mobility modes (walking and bicycling) changed with COVID-19-related motorized traffic reductions in Copenhagen, Denmark.

2. Methods

2.1. Study design

We opportunistically sampled particle number concentrations (PNC) from the onset of the COVID-19 societal closures implemented in Copenhagen in March to control the COVID-19 pandemic, Denmark. The local closures began mid-March 2020, with 'Phase 0' (March 13th, 2020). Our sampling began late-March (March 27th, 2020), in the middle of Phase 0, and continued through to Phase 3 (June 2020). See Table 1 for an outline of Phases, date ranges, and details of societal closures implemented. The societal closures starting in March coincided with reductions in total, and especially private vehicle, traffic counts in Denmark (Table 1). The lowest total counts during societal closures were recorded in week 13 of 2020, 43% lower compared to week 9 in the same year before implementation of the societal closures, after which traffic counts slowly increased again towards reaching pre-closure levels in June 2020 (Vejdirektoratet, 2021). All walking or bicycling trips were performed at approximately the same time of day over three months, between March and July, 2020 (from 'Phase 0' to 'Phase 3' societal re-opening).

Our chosen *walking* route is a lap around an inner-city lake, which is popular among both walkers and joggers. The south-west part of this route is close to streets with high traffic intensity of around 27,000 vehicles daily (pre-closure) (Teknik og Miljøforvaltningen/The Technical and Environmental Administration, Center for Trafik og Byliv/Centre of Traffic and Urban Life, 2015), while the path is further away from traffic in the north-east. The distance of this route is approximately 2 km. Monitoring of the route started within the time period of 12:30–14:30, once per day on weekdays. This time was chosen to cover a typical 'after-lunch-walk' around the lakes, which is popular among people working in offices in Copenhagen's city center.

Our chosen *bicycling* route is an inner-city lap passing by a regulatory curbside UFP monitoring station – see Supplemental Fig. S1. This route leads along streets with often high traffic intensity of up to 50,000 vehicles daily (pre-closure) (Teknik og Miljøforvaltningen/The Technical and Environmental Administration, Center for Trafik og Byliv/Centre of Traffic and Urban Life, 2015), as well as busy intersections and street canyons. The distance of this route is approximately 5 km. Monitoring of this route started within the typical afternoon commute time period of 15:30–17:30, once per day.

Either route was estimated to take approximately 20 min to complete (Fig. 1).

2.2. Personal exposure monitoring

A handheld nanoparticle counter ('DiSCmini'; Testo SE & Co. KGaA, Germany) was used to measure UFP at 1-second intervals. The instrument's impactor was connected to a flexible polymer sampling tube, connected to the instrument, and was fixed close to the bicyclist's/walker's collarbone to allow air sampling within the breathing zone. According to the manufacturer and previous studies (Ragettli et al., 2013; Luengo-Oroz and Reis, 2019) the DiSCmini measures particle number concentrations within the diameter range of 10–300 nanometers (nm), up to 1 million particles per cubic centimeter of air (pt/cm^3).

Table 1
COVID-19 phased closure and re-opening of Copenhagen, Denmark.

Phase	Date range of implementation	Details of implementation	Change in total traffic counts[a]	Number of monitoring days walking/cycling
0	13/03/2020–14/04/2020	Societal closures (closure of all non-essential services and workplaces)	−41% (week 12–15)	2/4
1	15/04/2020–10/05/2020	"First phase" of re-opening (essential services, including nurseries, kindergartens)	−25.8% (week 16–19)	10/9
2	11/05/2020–26/05/2020	"Second phase" of re-opening (non-essential services, including restaurants, cafes, etc.)	−13.6% (week 20–22)	8/9
2.1	27/05/2020–07/06/2020	"Second iteration of second phase" of re-opening (additional cultural and recreational activities, institutions of higher education, etc.)	−11.5% (week 22–23)	5/4
3	08/06/2020–21/07/2020 +	"Third phase" (wider re-opening of non-essential workplaces, including universities)	−4% (week 24–26)	16/17

Adapted from the Danish authorities' joint website on the COVID-19 outbreak in Denmark (Danish National Police, 2020).
[a] Changes in traffic counts are given as national, weekly values, compared to week 9, 2020, and are here averaged across phase periods (Vejdirektoratet, 2021). Only national data was available, which can be seen as indicative of trends in Copenhagen.

For quality control purposes, according to manufacturer recommendation, 'zero checks' were performed immediately before and after measurements using a HEPA filter. Further, validation of data was made by week-long co-location at a regulatory air quality monitoring station (H. C. Andersens Boulevard) at the beginning and end of the measurement campaign, in March and June. The station is equipped with a Scanning Mobility Particle Sizer (SMPS) that counts particles with mobility diameters between 11 and 478 nm every 3 min.

Concurrent geospatial coordinates were recorded with a GPS watch ('Forerunner 920XT'; Garmin Ltd., USA). Measurements were not collected on days or at times with precipitation or high humidity (>90%) according to the manufacturer's recommendation.

Background concentrations of UFP, coinciding with time of trip performance, were obtained from the regulatory air quality monitoring station (H. C. Andersens Boulevard, Fig. S1).

Meteorological information, as hourly means, coinciding with trip times, of temperature, relative humidity, wind speed and wind direction, was collected during the study period at a nearby monitoring station (H. C. Ørsted).

2.3. Statistical analysis

First, all time points where the HEPA filter was attached were excluded (33%/32% of total dataset for walking/cycling, respectively) and data with a particle diameter equal to zero (both 7% of total dataset) or PNC above 1 million (both <0.01% of total dataset) were removed for quality control purposes. PNC and diameter were aggregated as means per each trip time and merged with meteorological parameter means at the corresponding time. Next, PNC while walking or bicycling was described as mean, standard deviation (SD), median, range and interquartile range (IQR) of trips per transport mode.

For validation of UFP data, which was measured during bicycling, comparisons were made to a regulatory-grade particle counter within a regulatory air quality monitoring station that is located on the bicycling route (H.C. Andersens Boulevard). Trip means were compared with time-coinciding 20-minute means from the nearby station using Welch Two Sample t-tests. Further to this, with co-location of the DiSCmini at the same station for several weeks before and following our measurement campaign, we compared hourly means of either instrument using Spearman correlation tests and Bland Altman plots.

Linear models were used to examine the difference in PNC across COVID-19 re-opening phases. In a basic model, adjustment was made for time trend (a numeric variable of the date) and day of week using a Generalized Additive Model (GAM) (Wood and Scheipl, 2020). In adjusted models, re-opening phase, temperature, wind speed and wind direction were included one at a time. Furthermore, smoothing plots were used to visualize the relationship between PNC and meteorological factors. Degrees of freedom (df) for smoothing terms were determined based on unbiased risk estimation (UBRE) (Peng and Dominici, 2008).

Fully adjusted models were constructed as follows:

$$PNC\bar{s}(time\ trend, df = 9) + factor(day\ of\ week) + factor(phase)$$
$$+ factor(wind\ direction) + s(wind\ speed, df = 9)$$
$$+ s(ambient\ temperature, df = 9)$$

where:

time trend = numeric variable based on date;

phase = category 0, 1, 2, 2.1 and 3 based on phased societal re-opening dates;

wind direction = category 1, 2, 3, 4: 1–90, 91–180, 181–270, 271–360°;

s is for a smoothing term for time trend, wind speed or ambient temperature.

Medians of all trip data were geospatially aggregated in buffer zones of 20 m/30 m for bicycling/walking and visualized using ArcGIS (ESRI).

All statistical (excluding geospatial) analyses were performed in R (version 4.0.2). Statistical significance was accepted at p < 0.05.

3. Results

3.1. Data availability and validation

Approximately 100,000 s of data were collected in total, which were aggregated as means of 41 walking (n = 43,600) and 43 bicycling (n = 46,200) repeated trips. Table 2 describes particle exposure and meteorological parameters while either walking or bicycling. Supplementary material, Table S1, shows summary statistics of the total dataset.

At the beginning and end of these three months, the multiple week co-location period showed that data from the DiSCmini was comparable to a regulatory-grade air quality monitor. The DiSCmini measured 34% higher hourly mean PNC compared to the regulatory monitor at the beginning compared to 14% at the end. The correlation coefficient resulting from a Spearman test was 0.92 in the beginning compared to 0.71 at the end. See Supplementary material (Fig. S2) for Bland Altman plots.
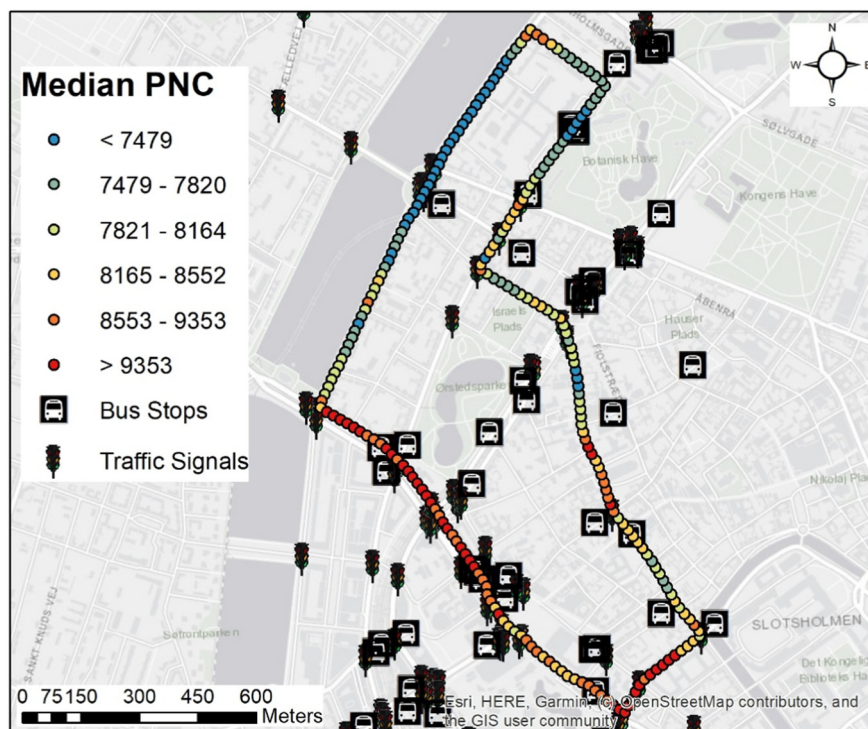
Coinciding 20-minute means from the time of bicycling trip performance were compared and seen as not significantly different between the DiSCmini and the regulatory monitor (p = 0.08; t = −1.79; df = 41.2; Fig. S3).

(a)
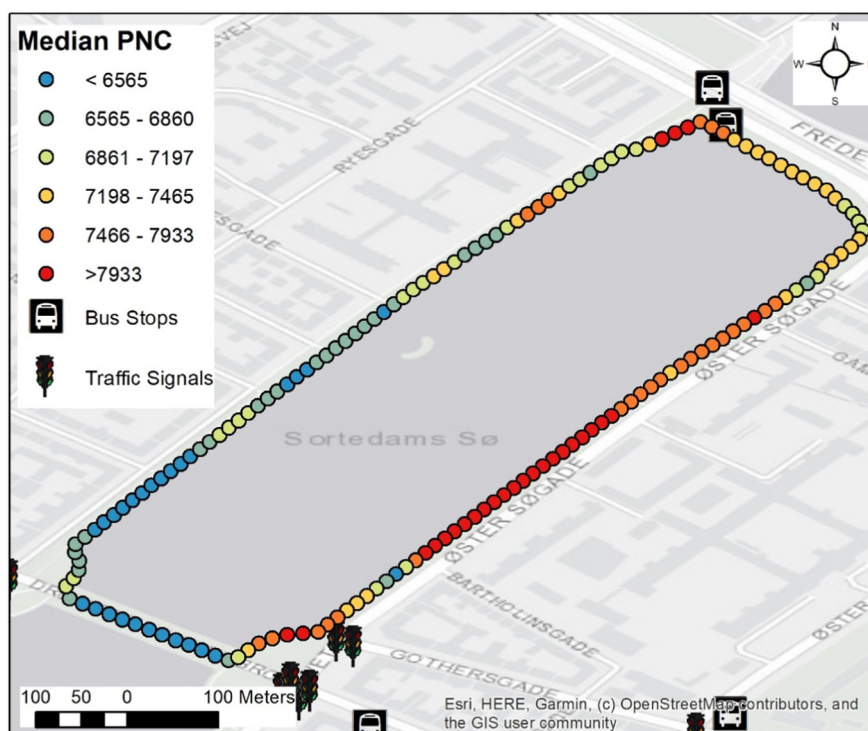
(b)

Fig. 1. Spatially aggregated median PNC while (a) walking and (b) bicycling. PNC data was spatially aggregated from all trips to show 'hot' and 'cold' spots along walking and bicycling routes of inner-city Copenhagen, Denmark.

3.2. Personal exposure levels

Mean and median exposure to UFP during trip times was lower for walking compared with bicycling, with mean exposures of 8810 particles per cubic centimeter of air (pt/cm³) while walking and 11,963 pt/cm³ while bicycling. Mean particle size was equivalent for walking and bicycling at approximately 40 nm (Table 2). Supplemental Table S1 shows summary statistics of the total dataset.

Geospatial aggregates of all trip data illustrated hot-spots of PNC along the route of either activity, largely along busier roads and intersections. For example, PNC hot-spots were shown close to intersections and traffic signals along Øster Søgade while either walking or bicycling, and along the entirety of H. C. Andersens Boulevard while bicycling (Fig. 1).

Across the study period, our repeated measures showed variation in PNC means across individual days for both walking and bicycling

Table 2
Description of trip means (all phases) of UFP exposure and meteorological parameters either walking or bicycling.

| Parameter	Mode	Mean	SD	Median	Min	Max
Particle number	Walking	8810	6827	6625	2368	41,329
(pt/cm³)	Bicycling	11,963	7297	9521	1239	33,661
Particle size (mean;	Walking	39	1.7	42	12	81
nm)	Bicycling	38	15	38	15	74
Temperature (°C)	Walking	16.2	5.7	16.5	6.8	27.0
	Bicycling	16.5	5.4	16.6	6.9	26.9
Wind speed (m/s)	Walking	5.0	1.7	5.0	2.4	8.3
	Bicycling	4.6	1.7	4.2	2.0	9.1

Abbreviations: N, sample size; nm, nanometer; m/s, meters per second; pt/cm³, particles per cubic centimeter of air; SD, standard deviation; min, minimum value; max, maximum value.

(Fig. 2). Across COVID-19 societal re-opening phases, our repeated measures showed statistically significant differences in PNC mean for bicycling, but not for walking (Fig. 3).

PNC while walking was highest at the start of the study period, during societal closure Phase 0, and decreased monotonically from Phase 0 to Phase 3 (Table 3). Similarly, PNC while bicycling was highest during Phase-0 and decreased until Phase 2.1, before it increased again in Phase 3.

3.3. Effect of covariates on personal exposure levels

Our adjustment for meteorological factors showed their significant influence on PNC. While the basic model with adjustment for time trend and day of week explained about 63% and 60% of deviance in PNC, respectively, adjustment for wind speed, temperature and wind direction combined added about 28% and 34% of deviance in PNC from
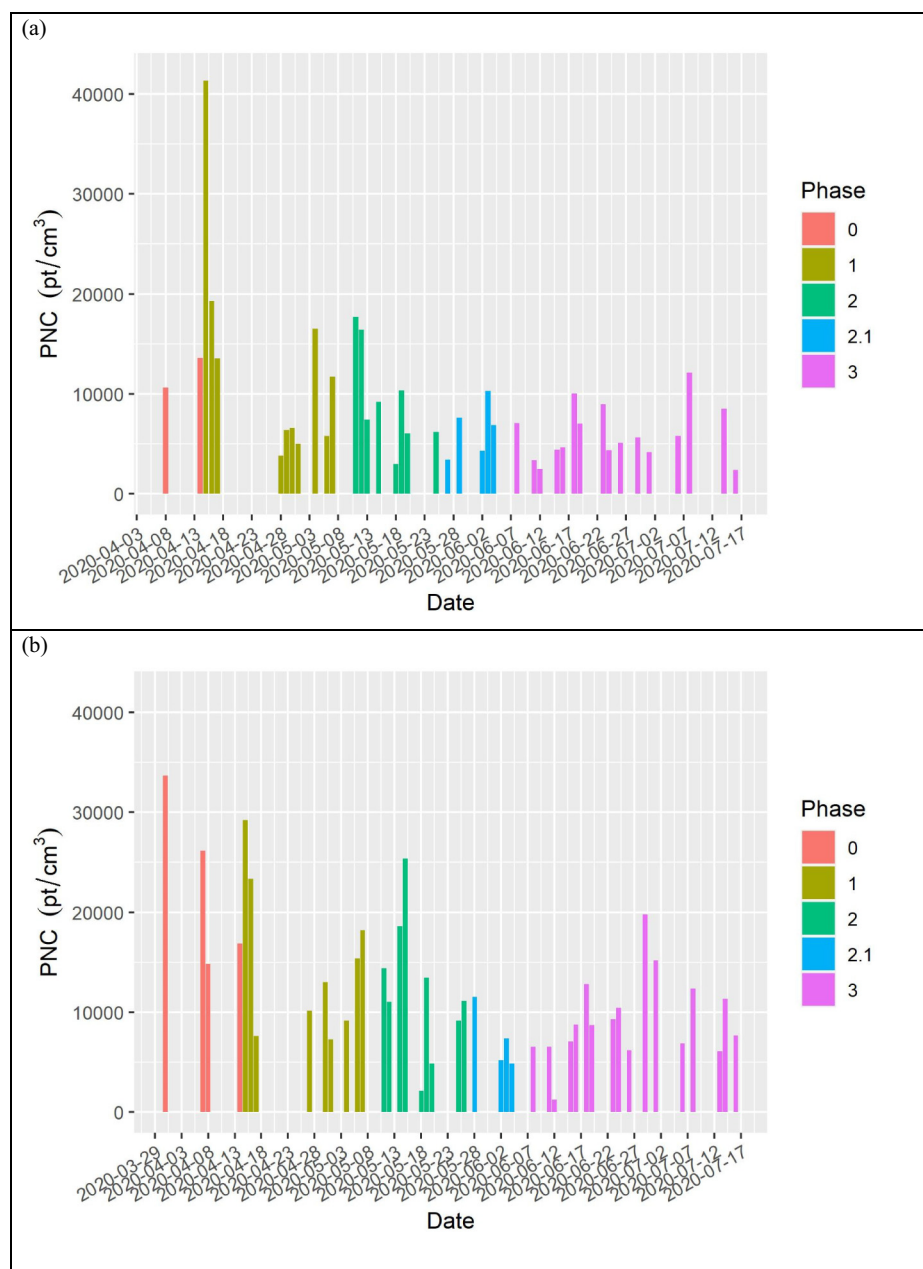
Fig. 2. Individual (a) walking and (b) bicycling trip means of PNC Trip (20-minute) means from measurements taken while walking or cycling, across individual dates from March to July.

(a)

(b)
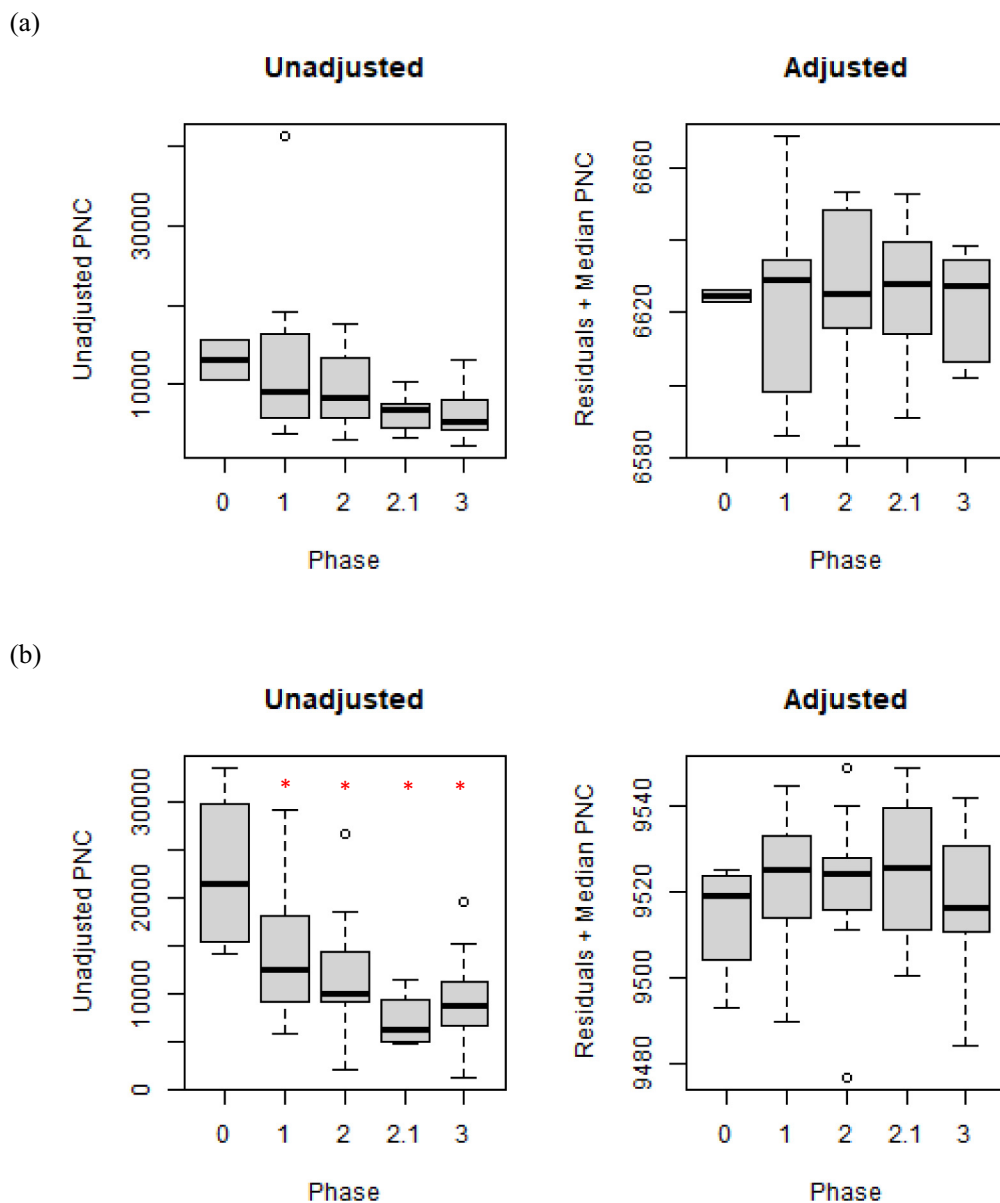
Fig. 3. Median and interquartile range of PNC across phases while (a) walking and (b) bicycling – adjusted and unadjusted for time trend, day of week and meteorological parameters. Abbreviations: PNC, particle number concentration. *Statistically significant (<0.05) change in PNC between phases. A GAM was used to adjust PNC across phases separately for walking or bicycling. The GAM equation used is as follows: PNC ~ s(time trend) + factor(phase) + factor(day of week) + factor(wind direction) + s(wind speed) + s(ambient temperature). The adjusted graph shows a boxplot of the median PNC across phases plus residuals from the GAM. The result is an explanation of the deviation in median PNC across phases without the effect of the time trend, day of week and meteorological parameters. Note different y-axis scales.

Table 3
Exposure levels of PNC while walking and bicycling across societal re-opening phases.

Phase	Mode	Mean	SD	Median	Min	Max	N of trips
0	Walking	13,170	3560	13,170	10,653	15,688	2
	Bicycling	22,672	8964	21,477	14,074	33,661	4
1	Walking	12,981	11,272	9153	3715	41,329	10
	Bicycling	14,519	7836	12,457	5781	29,246	9
2	Walking	9449	5169	8303	3090	17,572	8
	Bicycling	12,040	7407	9943	2140	26,806	9
2.1	Walking	6539	2705	6854	3429	10,296	5
	Bicycling	7241	3053	6297	4865	11,504	4
3	Walking	6068	2949	5367	2368	13,051	16
	Bicycling	9162	4309	8714	1239	19,614	17

Abbreviations: IQR, interquartile range; pt/cm^3, particles per cubic centimeter of air; Min, minimum value; Max, maximum value; N, sample size.

walking and bicycling, respectively (Table 4). When analyzed individually, wind speed had the strongest influence on PNC (walking: 14.4%, bicycling: 21.9%), followed by temperature (walking: 12.6%, bicycling: 10.4%). While the raw data shows statistically significant changes in PNC across phases for bicycling, these effects become insignificant after adjustment for covariates (Fig. 3 and Table S2). Smoothing plots for the effect of the separate meteorological parameters on PNC showed an inverse relationship between temperature and PNC, whereas wind speed had a less clear, but overall positive effect on PNC (Supplemental Fig. S4).

The levels and variability of meteorological factors, including temperature, wind speed, wind direction, and humidity, across phases are presented in Supplemental Fig. S5. Mean temperature increased from around 11 °C in Phase 0 to 21 °C in Phase 3, while wind speed decreased from 5 m/s to 4 m/s in the same period, respectively.

Table 4
Deviance of PNC exposure levels explained by different covariates while either walking or bicycling.

Covariate	Walking		Bicycling	
	Cumulative deviance explained (%)	Added percent points	Cumulative deviance explained	Added percent points
Time trend + day of week	62.5	Ref.	59.6	Ref.
+Re-opening phase	68.4	5.9	63.1	3.5
+Wind speed	82.8	14.4	85	21.9
+Temperature	95.4	12.6	95.4	10.4
+Wind direction	96	0.6	97.5	2.1

A GAM was used to determine the added deviance explained by adding individual covariates to the basic model of 'PNC ~ s(time trend) + factor(day of week).

4. Discussion

In this opportunistic repeated measures study, we found that phased re-opening of society and subsequent change in traffic intensity following COVID-19 closures in Copenhagen in the period of March to July of 2020 did not coincide with increases in UFP exposure while walking or bicycling. The observed gradual reduction in PNC during re-opening phases was to a large extent explained by meteorological parameters, namely wind speed and temperature, indicating that meteorology and seasonal variations were stronger drivers of UFP levels in this study period than COVID-19 societal closures and related changes in traffic counts. Moreover, we observed that exposure to UFP while bicycling was higher than while walking, with peak exposure to particle number concentrations being approximately double for bicycling compared to walking. This difference is probably related to differences in the chosen routes for walking and bicycling, as bicycling was performed closer to busy roads with motorized traffic, a main source of UFP. Along both routes, hotspots in UFP exposure were observed due to physical proximity and congestion associated with crossing of intersections (e.g., stopping at traffic signals), which we could visualize using geospatial information.

4.1. Comparison of our personal exposure levels to other studies

Several studies have measured UFP exposure while bicycling, mostly in Europe and North America. Among them, some studies compare exposure at different times of the day (Ragettli et al., 2013; Hofman et al., 2018; Berghmans et al., 2009; Kaur et al., 2005; Hatzopoulou et al., 2013; Qiu et al., 2019; Peters et al., 2014; Hankey and Marshall, 2015) or on different routes (high- vs. low-exposure traffic routes) within a city (Luengo-Oroz and Reis, 2019; Qiu et al., 2019; Hankey and Marshall, 2015; Pattinson et al., 2017; Cole-Hunter et al., 2012). Other studies have aimed at comparing exposure for different modes of transportation, such as car, bus, metro, bicycle or walking (Ragettli et al., 2013; de Nazelle et al., 2012; Strasser et al., 2018; Okokon et al., 2017; Zuurbier et al., 2010; Kingham et al., 2013; Kaur and Nieuwenhuijsen, 2009; Int Panis et al., 2010; Boogaard et al., 2009).

Six studies conducted in Belgium, Canada, Switzerland, China and the UK found higher UFP levels during morning rush-hours compared to afternoon rush-hours (Ragettli et al., 2013; Hofman et al., 2018; Berghmans et al., 2009; Kaur et al., 2005; Hatzopoulou et al., 2013; Qiu et al., 2019). Our observation of lower PNC while walking compared to bicycling could also be time-of-day dependent, with walking measured in the early afternoon (non-rush-hour) compared to bicycling in the afternoon rush-hour. Alternatively, other studies have found increasing levels of UFP in relation to closer proximity to traffic (Luengo-Oroz and Reis, 2019; Hankey and Marshall, 2015; Pattinson et al., 2017; Cole-Hunter et al., 2012). This would largely explain the same – lower PNC while walking, several meters further from road traffic, compared to higher PNC while bicycling. A review comparing UFP exposure in different modes of transportation found that car drivers tend to be most exposed to UFP, followed by people travelling by bus, bicycling and walking (de Nazelle et al., 2017).

There is only a single study that has quantified exposure to UFP during commute in rush-hour during weekdays in Copenhagen, in 2005 on a route comparable to ours, which reported an average exposure of 32,400 pt/cm³ (Vinzents et al., 2005), which is nearly three times higher than that observed in our study. This difference in levels since 2005 and 2020 may reflect improvements in fuel standards and an increasing share of emission-free and active modes of transportation over the 15 years.

4.2. Implications of meteorological factors on personal UFP exposure

Meteorological factors, mainly wind speed and temperature, have been shown to influence variation in air pollution levels between days and seasons of the year (Kaur et al., 2007; Knibbs et al., 2011). Both have previously been found to be inversely related to UFP levels – the increase in temperature and wind speed leads to lower UFP levels (Hatzopoulou et al., 2013; Peters et al., 2014; Thai et al., 2008). Wind direction can also have an influence on UFP levels (Hofman et al., 2018).

In our study, mean temperature showed an overall inverse relationship with mean PNC, as expected from previous studies. However, unexpectedly, mean wind speed showed an overall direct relationship with mean PNC. This unexpected finding could be due to the orientation of the routes, with either activity making a loop and meaning PNC measurements were made always both up- and down-wind of UFP emission sources. As such, when away from busy roads (motorized traffic), a strong wind could carry UFP emissions to the point of measurement, thereby increasing the measured concentration.

4.3. Implication/s of change in exposure levels during societal closures

Several studies have monitored the change in air quality due to COVID-19-related reductions in activity, such as closures of schools, workplaces, factories, and commuting traffic. The stringent societal closures to control the spread of the virus also decreased ambient air pollution levels including particulate matter ($PM_{2.5}$, PM_{10}), nitrogen dioxide (NO_2), and carbon monoxide (CO) – largely combustion or traffic-related emissions (Saadat et al., 2020; Muhammad et al., 2020). In Denmark, where Copenhagen is the capital, the national traffic counts for private cars and heavy duty vehicle traffic were reduced with 48% at the lowest level, and with a maximum reduction by 34% in only one week (Vejdirektoratet, 2021). A report based on preliminary data of $PM_{2.5}$ and NO_2 suggests a marked decrease in concentrations in 2020 compared to 2019 (Ellermann et al., 2021). Besides meteorological patterns and a general improvement in air quality in Denmark, this can at least partly be attributed to the COVID-19 societal closures. In March and April 2020, after introduction of COVID-19 restrictions, levels of NO_2 recorded at a street-level station in Copenhagen were as much as 40% lower than in the same period in 2019 and approached normal levels again as society was re-opened.

Like Denmark, the Republic of Korea did not impose strict measures to reduce or stop the movement or freedom of its citizens. After the sharp increases in COVID-19 cases in February 2020, the government of the Republic of Korea promoted "social distancing" by encouraging telecommuting and reduced outdoor activities, which eventually

resulted in a reduction of industrial operation and traffic volume. As a result, compared to air pollution levels in the previous (most recent) four years, a decrease of 45% for $PM_{2.5}$ and 20% for NO_2 were observed after 2020 social distancing in the Republic of Korea (Ju et al., 2020; Han and Hong, 2020).

Others found that $PM_{2.5}$ decreased by up to 30 μg/m^3 in major cities in China after the shutdown of industrial activities and transportation for approximately one month (Wang et al., 2020). Traffic-sourced PNC emissions were estimated to have reduced by more than 40% following societal closures in China (Dai et al., 2020). Other pollutants such as NO_2, highly correlated with UFP as a traffic-related pollutant, were estimated by satellite monitoring to decrease by up to 20% following this shutdown in eastern and central China (Chen et al., 2020; Bao and Zhang, 2020; Dutheil et al., 2020). One study of a neighborhood in the USA found up to 70% less traffic and up to an equivalent reduction in median PNC (Hudda et al., 2020). Another study investigating traffic-related air pollution, including UFP, while adjusting for meteorology found reductions in UFP significant only when making this adjustment (Xiang et al., 2020). They conclude that COVID-19 impacts on traffic-related air pollution levels can differ completely depending on weather scenarios, which means that our findings are not necessarily conflicting with theirs.

4.4. Strengths & limitations

Strengths of this study include repeated, real-time measurements using a benchmark personal particle counter sampling immediately within the breathing zone. Another strength is the verification of sampled personal exposure levels through the availability of 'background' regulatory monitoring stations, checking: (1) personal particle counter measurement accuracy per cycling trip, and; (2) personal particle counter measurement drift at the beginning and end of the multiple month-long study period per week-long co-location period. Importantly, we were able to make adjustments in our analyses for weather conditions per trip through the availability of corresponding weather station data.

This study has several limitations. Firstly, this study was opportunistic in design, initiated spontaneously from the onset of the COVID-19 societal closures. This opportunism meant that our power dictated by sample size is limited in its ability to detect differences in UFP exposure levels across phased societal re-opening. Unpredictable phase periods and staff availability or workplace access led to issues such as the low number of measurements in Phase 0. Secondly, the study would have been improved by comparing our findings to UFP levels measured at regulatory stations in the months before societal closures, or in previous years, in order to gain insights on seasonal trends in UFP levels. This was, however, not possible in the current study due to the regulatory data not being qualified for publication at the time of writing. Moreover, traffic data for this study was only available at a national level, and not specifically for Copenhagen. However, these trends can be seen as conservative of trends in Copenhagen, the capital and most populated city of Denmark. Third, we are aware of measurement drift that the DiSCmini may experience, meaning that instrument precision is questionable at higher compared to lower PNC and an offset may be required; to be formally investigated with future research. The manufacturer specifies that the DiSCmini is accurate for measuring PNC at ±30%, in agreement with studies that have compared the DiSCmini to regulatory-grade SMPS. These studies have found reasonably good agreement between instruments, suggesting that the DiSCmini is appropriate to use for personal exposure studies (Kaminski et al., 2013; Mills et al., 2013). Fourth, this study could not distinguish between different sources of UFP. While traffic is usually the main source of UFP in urban areas, other sources such as non-industrial combustion (e.g., wood stoves) also contribute to local UFP concentrations in Copenhagen, especially in residential areas (Jensen et al., 2013; Press-Kristensen and Råd, 2019). This may have influenced the higher levels of UFP in the beginning of the study period, coming out

of winter with people using wood stoves more frequently and spending more time than usual at home due to the COVID-19 societal closures, but also the lower levels of UFP going into summer with increasing temperatures. Finally, our study is novel and limited in representativeness as a commuting exposure study, outside of active mobility for physical activity purposes during work/school closures, to compare to previous commuting exposure studies. The bicycling route is unrealistic as a commute in that it is a loop, however it is comprised of popular paths and indicates exposure levels while performing active mobility. Obtaining physical activity through active mobility is particularly popular in Copenhagen, where 49% of inhabitants bicycled for their daily commutes to work or education in 2018; increasing from 38% in 2009 (City of Copenhagen, Technical and Environmental Administration, (TMF), Mobility, 2019). As physical activity is encouraged to promote health benefits including a strengthened immune system, active mobility can be promoted recreationally (e.g., walking or cycling in a loop) to maintain levels no longer achieved through daily commuting.

5. Conclusion

We observed decreases in UFP exposure during walking and bicycling commutes during societal re-opening phases related to the COVID-19 pandemic in the period from late March to mid July 2020 in Copenhagen, Denmark. This decrease was to a large extent explained by meteorological factors (wind speed and temperature), which seemed to be stronger drivers of UFP levels during this period than COVID-19 societal closures.

Future work will investigate peak versus off-peak commuting (with new city UFP monitoring stations), as well as accuracy drift of the DiSCmini device.

CRediT authorship contribution statement

M.L. Bergmann: Formal analysis, Data curation, Writing – original draft, Visualization. **Z.J. Andersen:** Conceptualization, Writing – review & editing, Supervision. **H. Amini:** Writing – review & editing, Visualization. **T. Ellermann:** Writing – review & editing, Resources, Data curation. **O. Hertel:** Conceptualization, Writing – review & editing. **Y.H. Lim:** Formal analysis, Writing – review & editing. **S. Loft:** Conceptualization, Writing – review & editing. **A. Mehta:** Conceptualization, Writing – review & editing. **R.G. Westendorp:** Conceptualization, Writing – review & editing. **T. Cole-Hunter:** Project administration, Conceptualization, Methodology, Writing – original draft, Data curation, Investigation.

Declaration of competing interest

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

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2021.148301.

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