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Research article

Particle number and mass exposure concentrations by commuter transport modes in Milan, Italy

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Abstract: There is increasing awareness amongst the general public about exposure to atmospheric pollution while travelling in urban areas especially when taking active travelling modes such as walking and cycling. This study presents a comparative investigation of ultrafine particles (UFP), PM10, PM2.5, PM1 exposure levels associated with four transport modes (i.e., walking, cycling, car, and subway) in the city of Milan measured by means of portable instruments. Significant differences in particle exposure between transport modes were found. The subway mode was characterized by the highest PM mass concentrations: PM10, PM2.5, PM1 subway levels were respectively about 2-4-3 times higher than those of the car and open air active modes (i.e. cycling and walking). Conversely, these latter modes displayed the highest UFP levels about 2 to 3 times higher than the subway and car modes, highlighting the influence of direct traffic emissions. The car mode (closed windows, air conditioning and air recirculation on) reported the lowest PM and UFP concentration levels. In particular, the open-air/car average concentration ratio varied from about 2 for UFP up to 4 for PM1 and 6 for PM10 and PM2.5, showing differences that increase with increasing particle size. This work points out that active mode travelling in Milan city centre in summertime results in higher exposure levels than the car mode. Walkers' and cyclists' exposure levels is expected to be even higher during wintertime, due to the higher ambient PM and UFP concentration. Interventions intended to re-design the urban mobility should therefore include dedicated routes in order to limit their exposure to PM and UFP by increasing their distance from road traffic.

Keywords: exposure concentration; transport mode; ultrafine particle; particulate matter, active mobility

Abbreviations:

CV: coefficient of variation

PM: particulate matter

PNSD: particle number size distribution

ME: microenvironment

PNC: particle number concentration

UFP: ultrafine particles (dp < 100 nm)

1. Introduction

Long-term exposure to fine particulate air pollution is associated with non-accidental cause mortality, even within concentration ranges well below the European annual mean limit value [1]. In addition, an increasing number of both epidemiological and toxicological studies suggest a great potential for ultrafine particles (UFP - $d_p < 0.1$ µm) to affect human health [2,3]. Most of the documented associations were derived by time series analysis, relating day-to-day variation of air pollution and mortality and morbidity data [4]. In these analyses the population exposure to particles was usually assessed indirectly based on outdoor concentrations measured at fixed monitoring stations. However, it is widely recognized that, because people spend most of their life indoors and in transit, the daily exposure is largely determined by the particle concentration that people experience in these microenvironments (MEs, i.e. home, school, workplace, transport microenvironments), where the concentrations can be significantly different from outdoors [5]. In particular, exposures to high average levels and to short-duration concentration peaks of fine and ultrafine particles may occur in transport MEs because of the proximity to the emission sources [6]. So far, different studies have focused their attention on transport MEs directly measuring personal exposure to various size fractions of PM mass (PM10, PM2.5, PM1) and more recently to UFP number concentration by means of portable instruments. Karanasiou et al. (2014) [7] reviewed several European studies on personal exposure during commuting by different modes of transports and suggested that the commuter exposure to traffic related pollutants (PM, black carbon, UFP in particular) may be potentially reduced by a forward-looking, integrated transport policy, involving the phased renovation of existing public vehicles and the withdrawal of the more polluting (older) private vehicles, combined with incentives to use public transport and the encouragement of commuter physical exercise.

A number of studies directly compared the exposure concentrations, i.e.: the concentrations to which a person is exposed (hereafter referred to as exposure levels), among different transport modes [8-10]. The available literature reports in some cases contrasting results. For instance, different studies have observed lower exposure levels to both UFP and PM for walkers and cyclists than for car and bus passengers [11]. However, some authors [12,13] have found an opposite result and according to another study [14] cyclists and car passengers experience similar UFP exposure. Reductions in cyclists' exposure have been observed when they take alternative routes along lower trafficked roads [15-17]. Conversely, studies comparing cyclists' and drivers' exposure to CO and other gaseous pollutants consistently report increased exposure when driving [11,17-19]. These contrasts highlight the dependency of the exposure levels on a large number of variables such as road characteristics, meteorological conditions, vehicle cabin ventilation/conditioning system, and vehicle fuel [10,20].

Some other studies considered the dose experienced by the commuters, thus accounting for mode-specific inhalation rates, exposures, and trip duration [21,22]. Indeed, the minute ventilation is higher for the active commuting modes (i.e. walking and biking) as the physical effort results in both higher breathing frequency and larger tidal volume [23]. Compared with passive modes, ventilation rate is reported to be up to 5 times higher for biking and about 3 times for walking [14,24]. Additionally, the lung deposition rate of inhaled particles is strongly increasing with exercise [25,26], moreover favoring the deposition in the lower airways. Therefore, active commuting can result in higher particle deposition in the alveolar region.

In Italy, and in particular in Milan, a well-known European hot-spot for PM pollution, very few field experiments [6,27,28] have been conducted studying both particle number concentration (PNC) and particle mass in different commuter modes. In order to address this gap, this paper investigates the impact of urban transport mode choice on commuters' exposure to airborne particulate matter based on field measurements performed while travelling an assigned route in the city centre of Milan by four different transport modes (i.e. walking, cycling, car, subway). While the main focus of this work is on UFPs, information on PM10, PM2.5, PM1 mass concentration levels derived from PNC data is also discussed. Comparisons between the particle mass and number exposure levels by travel mode are presented and an exploratory cluster analysis is applied in order to investigate differences and similarities between PNCs characterizing the four transport modes.

Due to the serious problems in complying with the European Union air quality standards for both PM10 and PM2.5, the municipality of Milan has introduced a congestion charging scheme to decrease access to the city centre, reduce congestion, improve public transport networks and to increase the modal share of sustainable transport modes (car pooling incentives, and public bike sharing service). The traffic restricted area and the bike sharing services are expected to expand to a larger area. However, there is a growing concern of public opinion about exposure to atmospheric pollution while travelling in the urban area especially when taking active travelling modes. Therefore, this work is intended to provide a first knowledge on the particle exposure levels for the most popular transport modes, used daily by thousands of residents and commuters in Milan city, outside the congestion charging scheme area, suitable for actual exposure assessments.

2. Materials and Methods

2.1. Instrumentation

The personal exposure to PNCs was measured by means of a portable apparatus equipped with two aerosol monitors: a condensation particle counter (P-Trak, TSI Model 8525, USA) and an optical particle counter (OPC - Personal DustMonit, Contec, Italy). Both instruments were held in a backpack during the monitoring campaigns.

The P-Trak counter is a portable condensation particle counter able to measure in real-time the PNC in the 20–1000 nm size range ($PNC_{20–1000}$) at 1-min time resolution, detecting particle concentrations up to 5×10^5 cm^{-3}. Ambient air drawn into the instrument is first saturated with isopropyl alcohol vapor that then condenses onto the particles, causing them to grow into a larger droplet detectable by means of a photo-detector when flashed by a focused laser beam. Despite its measurement range extending beyond 100 nm, P-Trak data are commonly regarded as UFPs concentration data, since in urban areas particles with diameter below 100 nm account for the majority of the total particle number; therefore, hereafter $PNC_{20–1000}$ data are presented as UFP data.

The Personal DustMonit OPC measures the PNC for 7 size bins in the 0.3–10 µm size range at 1-min time resolution by means of laser light scattering technology. Ambient air is drawn into the measurement cell at a rate of 1 liter per minute by means of a straight sampling tube (7 mm internal diameter, 40 cm length) with the inlet located in the breathing zone. Due to the straight configuration of the sampling line no significant particle losses are expected in the sampling tubing. Size classified PM mass concentrations were separately calculated for each of the 7 size bins from PNC data by means of corresponding size-resolved conversion factors k_i (kg m^{-3}) under the assumption of spherical shape of the particles. These conversion factors $k_{i,}$ specifically estimated by multiple linear regression technique based on data from collocated parallel field tests of the OPC instrument and PM10 and PM2.5 gravimetric measurements in Milan [29], actually account for the particle density, for a shape factor considering non-sphericity, and for the cut-off efficiency of the gravimetric samplers (EN 12341 and EN 14907 reference methods) since the OPC has no size-selecive inlet for particles. For the subway ME, where an important PM enrichment in heavy metals such as iron resulting from the wear of wheels, rails and brakes is expected [30], the conversion factors have been derived based on concurrent gravimetric PM data and PNC data collected in the Milan subway system by least square regression technique [31].

The size classified PM mass concentration data were then summed up in order to obtain the PM mass of dimensional fractions of interest in this study, namely $PM_{0.3–10}$, $PM_{0.3–2.5}$, and $PM_{0.3–1}$. However, due to the negligible contribution in terms of mass from particles below 0.3 µm, these latter mass fractions can be reasonably regarded as PM10, PM2.5, and PM1.

2.2. Monitoring locations and design

The monitoring campaigns were carried out in Milan city centre, outside the congestion charging scheme area, during two weeks in July 2010, on three workdays per week. Four transport modes, representing the most popular commuting ways in Milan city area, were investigated: three surface transport modes (i.e. walking, cycling, car) and one underground mode (i.e. subway). Data were collected on a 2-km long route along a city-centre trafficked 4-lane road (Figure 1), characterized by a daily average traffic intensity of 2–2.5 thousands vehicles per hour, mainly consisting of passenger cars and mopeds, but also inlcuding a 15% of light duty vehicles. The road, bordered by four-, five-story buildings, is a major road axis leading to city centre from North-East; additionally, it is one of the most crowded areas for shopping in Milan. The M1 subway line runs in the same direction under the road, with the top of rail about 10 m below ground level. The selected route was travelled consecutively with each of the four different transport modes. Each single trip lasted about 30 minutes, allowing 1 round trip (back and forth) for the walking mode and 2 round trips for all the other modes, for a total of a 2-hour campaign per day. The car used for the campaigns was a Ford Focus S-MAX (2008) petrol-fueled and equipped with an air filtration system. During all car trips, the car windows were closed, the air conditioning was turned on, set on cool air and moderate ventilation. These standard conditions were chosen because they are typical of the summer season. Before each car journey, the car cabin was fully ventilated maintaining doors open and ventilation system on set at maximum level. While the backpack containing the instruments was placed on the front passenger seat in the car, an operator wore the backpack keeping the instrument inlets near the breathing zone during the other travel modes.

The bike trips were performed along the right-most lane of the road since a dedicated bicycle lane was not present. Walking trips were carried out on the sidewalk, at a distance from the roadway of approximately 2 m. Subway trips were underground for the entire duration, including the time spent for short pedestrian journeys in subway stations, waiting on the platform and traveling in the train cabin. No distinction between mechanically and naturally ventilated train cabin was considered.

Figure 1. Map of the sampling route travelled with the different transport modes. Stars are traffic light regulated crossings; M squares the subway stations.

Travels with different transport modes were not simultaneous but were performed consecutively between 3 pm and 5 pm in the same order each time (i.e. car, bike, walk, subway). No significant variations in the meteorological parameters and in traffic flows occurred during the sampling days and hours. The nearest meteorological and urban air quality station operated by the local environmental protection agency recorded the typical summer conditions for Milan: ambient temperature around 30 °C, relative humidity around 45%, and ground-level wind speed around 1 m s^{-1} (Table 1). Ambient air pollution concentrations were also on usual levels with PM2.5 daily averages (the only data available) in the 8–20 μg m^{-3} range. Traffic flows during the trips for on-road modes were quite similar, as in Milan the summertime evening peak of traffic usually occurs after 6 pm. Even though hourly traffic counts were not available, according to the modeled traffic pattern for summer workdays, traffic intensity between 3 pm and 5 pm was fairly constant and in the orders of 3.4 thousands vehicles per hour. Therefore, despite not concurrent, it can be reasonably assumed that the different mode trips have been carried out under the same environmental and traffic conditions.

Table 1. Meteorological conditions during the monitoring campaigns.

Date	Time	T	Relative humidity	Pressure	Radiation	Wind speed	Wind direction
		(°C)	(%)	(kPa)	(W m^{-2})	(m s^{-1})	(Degrees)
day 1	3:00 PM	29.5	48	100.64	484	0.7	121
	4:00 PM	30.1	49	100.59	534	1.1	182
	5:00 PM	30.5	41	100.58	398	1.3	290
day 2	3:00 PM	28.5	43	100.97	795	1.2	160
	4:00 PM	29	43	100.92	670	1.1	146
	5:00 PM	28.8	44	100.9	512	1	122
day 3	3:00 PM	27.8	44	101.31	662	0.8	101
	4:00 PM	28	44	101.27	590	0.8	126
	5:00 PM	27.8	44	101.24	417	0.6	129
day 4	3:00 PM	30.7	47	100.5	751	0.8	139
	4:00 PM	31.1	46	100.45	642	0.8	184
	5:00 PM	31.2	44	100.39	474	0.5	126
day 5	3:00 PM	30.7	49	100.51	772	1.4	220
	4:00 PM	31.5	46	100.5	668	1.6	258
	5:00 PM	31.8	44	100.48	509	1.3	203
day 6	3:00 PM	32	46	100.77	730	0.8	128
	4:00 PM	32.4	44	100.74	666	0.9	133
	5:00 PM	32.6	39	100.7	521	0.6	156

2.3. Data processing

Recorded concentrations were analysed in terms of the main statistical parameters of the data distributions and of the correlation between mass and number concentration data for each transport mode separately. Representative size distributions of the PNC in the 0.3–10 µm range were derived from OPC data for each transport mode. One-way ANOVA analysis was applied in order to investigate whether statistically significant differences exist in particle exposure levels between transport modes. Homogeneity of variance test was run according to Levene's statistic prior to the application of the ANOVA. F-ratio tests was used when the homogeneity of variance was satisfied while Welch test when it was not. In all analyses a level of significance of 0.05 was assumed.

An unsupervised cluster analysis was then applied to the entire dataset of 1-min observations in order to investigate whether the four transport modes were discernible on the basis of the measured PNCs. The analysis was performed according to the k-mean clustering algorithm initialized to partition the original dataset into four clusters. The k-mean algorithm identifies the clusters by minimizing the sum of squared Euclidean distances of each observation from its cluster centroid using an iterative procedure. The input matrix of the clustering algorithm was based on the actual measured data (i.e. PNC data) and included as variables the 1-min PNC data in four size bins: UFP from P-Trak readings, $PNC_{0.3–1}$, $PNC_{1–2.5}$, and $PNC_{2.5–10}$ from OPC readings. In order to prevent the possibility that the different scales of the input variables could affect the clustering procedure, z-score standardization was applied prior to the cluster analysis, based on the mean and standard deviation of each variable. After clustering, average PNC and PM mass concentrations characterizing each of the four clusters identified by the algorithm were calculated and the occurrences of transport mode observations in the four clusters were studied.

3. Results and discussion

3.1. Particle number concentration

The summary statistics of 1-min UFP number concentrations for each transport mode are reported in Table 2 and plotted in Figure 2. On average, concentrations for the two open-air modes (bike: 2.9×10^4 cm^{-3}; walk: 2.2×10^4 cm^{-3}) were about 2–3 times higher than those for the subway (1.3×10^4 cm^{-3}) and car mode (1.1×10^4 cm^{-3}) that exhibited comparable average concentrations. Differences between modes are indicated as statistically significant by the ANOVA analysis. PNC levels measured for the open-air modes are in agreement with those reported for urban environments more or less directly influenced by traffic emissions [32,33]. Mean cyclists' exposure levels in the 1.6×10^4–2.8×10^4 cm^{-3} range are reported in studies developed in Switzerland, Belgium and The Netherlands [9,14,15,34]; much higher levels (4.5×10^4–8.4×10^4 cm^{-3}) have been reported in other Dutch studies and in Spain [11,16,35]. Car levels in this study are significantly lower than the average PNCs reported in other European studies but they are in rather good agreement with those reported by Spinazzè et al. (2015) [6] for car trips in Como, a mid-sized city 50 km North of Milan ($0.9 \pm 0.9 \times 10^4$ cm^{-3}). Subway concentrations were about 3 times lower than those reported in previous studies for subway rides in Milan [36] and 2 times lower than those reported for subway commuting in Barcelona [37].

All the surface modes displayed a large dispersion, as shown by the coefficients of variation (CV = 0.5, 0.6 and 1.1 for walk, bike and car mode, respectively), with extreme values (95th percentile) in the 3.4×10^4 cm^{-3} (car)–5.5×10^4 cm^{-3} (bike) range and maximum values ranging between 7.1×10^4 cm^{-3} (car)–10.3×10^4 cm^{-3} (bike). Conversely, the subway mode was characterized by a narrower data distribution (CV = 0.3) with extreme and maximum values in the orders of 1.9×10^4 cm^{-3}–2.2×10^4 cm^{-3}. The greater dispersion observed in the bike, walk, and car mode data arose mostly from intra-day variability rather than from day-to-day variability. In fact, high peaks in the order of 4–8 × 10^4 cm^{-3} repeatedly occurred in all the surface mode trips, typically in correspondence with walk stops or bike/car queuing at the traffic lights (Supplementary material, Figure S1). Differently, the UFP concentrations measured during subway trips appear more uniform and relatively stable.

The particle number size distribution (PNSD) was investigated based on OPC data. Even though limited to the 0.3–10 µm size range, this analysis pointed out interesting features of PM exposure corresponding to various transport modes.

Table 2. Summary statistics of 1-min UFPs number and PM mass concentrations for the four transport modes.

Statistical parameter	UFP (10^4 cm^{-3})				PM10 (μg m^{-3})			
	C	B	W	S	C	B	W	S
Mean	1.1	2.9	2.2	1.3	13.4	84.8	79.3	147.7
St. Deviation	1.2	1.7	1.0	0.4	16.0	48.1	49.9	91.3
Median	0.4	2.5	2.0	1.4	7.4	74.1	66.4	128.9
IQR	1.5	1.7	1.1	0.6	11.9	41.1	29.8	91.2
Min	0.1	0.6	0.6	0.5	0.8	35.5	36.9	34.8
Max	7.1	10.3	7.9	2.2	95.8	442.7	483.6	508.2
5th percentile	0.1	1.0	1.0	0.6	2.4	44.8	46.2	52.8
95th percentile	3.4	5.5	4.4	1.9	52.4	136.2	131.3	347
	PM2.5 (μg m^{-3})				PM1 (μg m^{-3})			
	C	B	W	S	C	B	W	S
Mean	3.9	23.1	20.2	91.1	2.7	12	10.7	36.7
St. Deviation	3.9	7.1	5.1	39.1	2.6	3.3	2.4	11.7
Median	2.4	22	19.9	82.8	1.5	12.3	10.5	35.8
IQR	4.4	9.2	5.5	42.1	3.0	5.6	3.1	16.8
Min	0.3	13.2	12.5	26.4	0.1	6.5	7.0	16.1
Max	26.8	64.9	48.1	204.5	16.7	20.2	22.2	74.7
5th percentile	0.5	15.2	13.9	36.3	0.2	7.2	7.6	20.1
95th percentile	9.2	33	26.8	171.1	6.7	17.3	14.9	56.7

C = car; B = bike; W = walk; S = subway

For all the transport modes, the size distribution displayed a bimodal shape resulting from the superposition of an accumulation mode, of which only the upper tail was observable, and a fine mode (Figure 3 and Supplementary material, Figure S2); the contributions of these particle modes to the size distribution were however rather different. The bike and walk PNSD, almost coincident over the entire size range, were characterized by a strong contribution from accumulation mode particles, with 94% of particles belonging to the finest size bin (0.3–0.5 µm) and a less relevant contribution from the second mode located in the 2–3 µm range.

The shape of the PNSD for the car mode was rather similar to those of bike and walk mode, but with concentration levels in each size bin significantly lower than the open-air modes: in particular, particles were from about 75% (finest size bin: 0.3–0.5 µm) up to 90% (0.7–3 µm size range) less than at open-air modes. Consequently, for the car mode the 96% of the particles belong to the finest size bin (0.3–0.5 µm). The PNSD of the subway differs from the bike and walk PNSDs, with a higher presence of particles in the size bins covering the range 0.5–3 µm. In particular, the particle number concentrations in the size intervals around 1 µm are about 4–5 times higher than those of the open-air modes, whereas similar concentrations are observed for the finest (0.3–0.5 µm) and largest (5–10 µm) size intervals. Thus, for the subway mode, the relevance of the finest size bins is reduced compared to the other transport modes, accounting for only the 80% of the total number concentration of particles in the size range 0.3–10 µm.

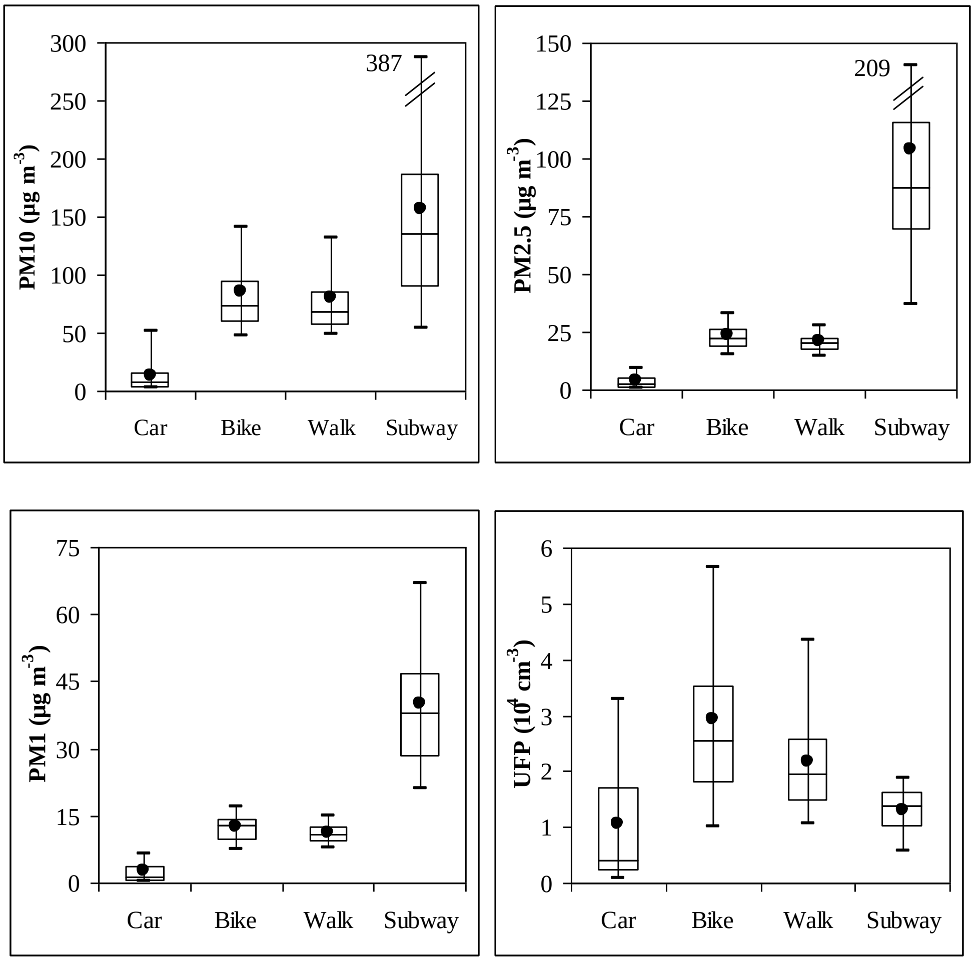

Figure 2. Box-plots for PM10, PM2.5, PM1 and UFP 1-min concentration data for transport modes.

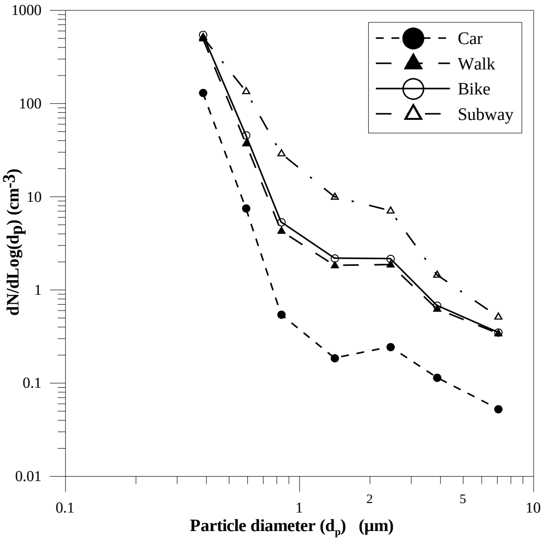

Figure 3. Average particle number size distributions (0.3–10 μm size range) for transport modes. (y-axis reports the size-resolved particle number concentration N normalized for the width of the corresponding size bin).

3.2. Particle mass concentration: PM10, PM2.5, PM1

The summary statistics of the distribution of the 1-min PM mass concentrations derived from OPC data are reported in Table 2 and represented in the box-plots of Figure 2, separately for each transport mode. Considerable differences in PM10, PM2.5 and PM1 levels across modes can be observed: while the two open-air modes display comparable PM levels, though slightly higher for bike than walk mode, the car and the subway mode display lower and higher concentration levels. The ANOVA analysis confirms the existence of statistically significant differences in PM10, PM2.5, PM1 averages between modes. According to the different size distributions that characterize the car mode, the difference in PM levels with respect to the open-air modes is higher for the larger size cuts, with a car/open-air average concentration ratio increasing from about 0.17 for PM10 and PM2.5 up to 0.24 for PM1. Inside-to-outside car ratios as low as 0.1 have been reported for PM2.5 and particle-related concentrations, depending on car ventilation settings [38]. PM1 concentrations about 50% lower than on-road concentrations have been observed in taxi trips with closed windows in Shanghai [24]. On the other hand, the subway mode displays the highest average exposure levels for PM10, PM2.5 and PM1, all about 2–3 times greater than those of open-air modes, as well as a wider range, especially for PM2.5 and PM1. In fact, a strong variability characterizes the 1-min PM mass concentrations during subway trips with high peaks of PM10, PM2.5 and PM1 repeatedly occurring in the subway ME (Supplementary material, Figure S3).

Coherently with the observed differences in the PNSDs, the subway/open-air average concentration ratio is larger for PM2.5 (4.2) and PM1 (3.2) than for PM10 (1.8). Furthermore, the difference between subway and open-air modes is also enlarged by the different nature of the particles, which strongly affects the k_i factors used to convert particle number concentrations into mass concentrations. In particular, the experimentally determined k_i factors, which incorporate the true density of the particles, exhibit larger differences in the fine super micron size range, where they are in the orders of 3.4 g cm^{-3} for the subway environment and about 2.7 g cm^{-3} outdoors [31].

3.3. Cluster analysis

The k-mean clustering algorithm, initialized to partition the entire dataset into four clusters, identified four well separated clusters, both in terms of particle concentration levels (Table 3) and in terms of transport mode occurrences (Figure 4). Cluster 1 displayed the highest average PM concentrations and was mostly constituted (92%) by subway observations, with some bike and walk mode data accounting for the remaining 8%; 40% of the total subway data fell in this cluster. In contrast, Cluster 4 was characterized by the lowest average concentration levels for both PM and UFP. Car mode data constituted the majority (96%) of cluster members, the remaining 4% given by some subway mode data and few walk data; 87% of car mode data fell in this cluster. Cluster 2 and Cluster 3 were mainly composed of data from the two open-air transport mode data, altogether accounting for 70% and 83% of the cluster populations, respectively. Cluster 2, which included the most part of bike (60%) and walk (80%) observations and the remaining subway and car data, was characterized by higher PM mass average levels than Cluster 3 and resembled the typical open-air conditions. These conditions, however, can be found also in those sectors of the subway ME (i.e., stairs to the intermediate underground level, ticketing halls) not directly influenced by train movements. A previous study on Milan subway stations showed that daytime PM10 concentrations at the mezzanine levels are in between those found in ambient air and at platform level, as a result of the mixing of ambient and platform level air [31]. Cluster 3, which reported the highest average UFP concentration levels, contained no subway data and the three other transportation modes showed almost the same frequency. Cluster 3 grouped those (less frequent) situations of high UFP peaks, observed only for the surface transport modes, typically occurring at road crossings where the accelerating vehicles from idling queues resulted in sudden bursts of UFP emissions, able to penetrate into the car cabin too.

Table 3. Cluster average concentration levels.

Cluster	PM10 (μg m^{-3})	PM2.5 (μg m^{-3})	PM1 (μg m^{-3})	UFP (cm^{-3})
1	227	123	41	1.4×10^4
2	88	36	18	1.6×10^4
3	61	17	9	4.0×10^4
4	13	4	3	0.8×10^4

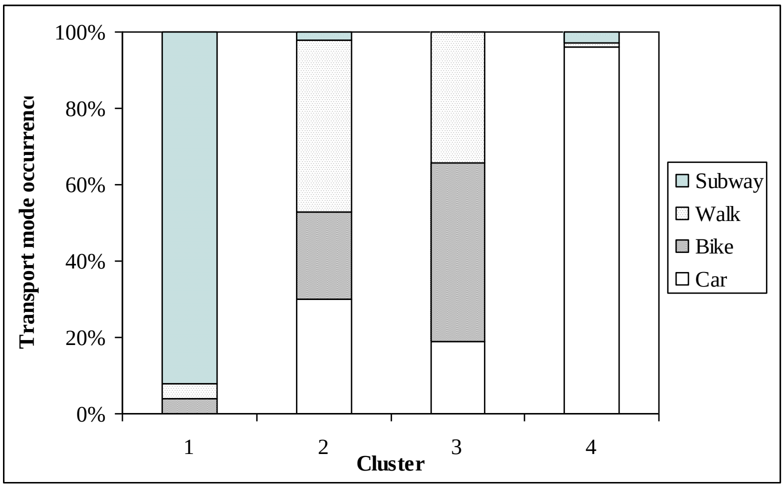

Figure 4. Occurrences of transport mode observations in the four clusters identified by the k-mean clustering algorithm.

3.4. Discussion

In this study, cyclists and pedestrians in Milan appear to be exposed to higher concentrations of both PM mass and particle number than car passengers. Though a number of studies reported opposite results [11,16,18,20], higher exposure levels for active mode commuting have been reported in some other studies [8,12-14,17]. Interestingly, the study in the nearby city of Como reported lower in-car exposure levels compared to active modes on both low-traffic and high traffic routes [6]. Among all the factors reported to affect exposure levels [10,20], in our case the road features combined with the meteorological conditions and the features of the test car are the most relevant. While the former factors acted in increasing the ambient concentrations, the latter contributed to reduce in-car concentrations; overall, these opposite effects enhanced the contrast between the travel modes. Indeed, the low speed of the wind, mostly blowing crosswind to the road, resulted in a weak dispersion of the local traffic emissions, furthermore limited by the urban canyon configuration of the road, thus favoring the build-up of ambient concentrations with a straightforward negative impact on the exposure levels for the active modes. On the other hand, the largely lower PM10, PM2.5, and PM1 concentrations that characterized the car mode with respect to the open-air modes are likely related to the insulating capacity of the air ventilation/filtration system from the outdoor environment. Studies where modern, medium-level cars were used and closed window and air recirculation conditions were set showed lower exposure levels for car commuters [6,8,17]. On the contrary, when utility cars [9,36] or open window settings [8,11] were used, car cabin concentrations were higher than outdoors. As supported by some further studies [39,40], design and combined filtration/ventilation system of newer cars results in cleaner in-cabin environment.

In particular, the filtration system of the car used in this study was more effective in abating the super-micron size fractions. In fact, the comparison between open air modes and car cabin PNSDs points out that the lowest reduction of PNC levels occurs for particles in the smallest size range (0.3–0.5 µm). Furthermore, the less relevant effect of the ventilation/filtration system on smaller particles is also suggested by the similar variability of UFP number concentration measured in all the surface modes.

The subway passengers in Milan appear to be exposed to higher PM concentrations than cyclists, pedestrians and car passengers, in agreement with studies reporting PM values much higher than outdoor levels in the subway system of large cities worldwide with average PM10 levels on platforms up to 300 mg m^{-3} [41, and references therein]. The subway ME was characterized by high and widely dispersed levels of PM concentrations and relatively low and constant levels of UFP number concentrations (Figure 2). Previous studies in subway ME reported several reasons for fluctuations in both PNC and PM levels, including station depth and design, ventilation system, train piston effect, train speed and frequency, wheel materials and braking mechanisms [30,42,43].

In this study, the major PM peaks most frequently occurred while waiting on the platform and travelling in old, naturally ventilated train cabins. Indeed, there was evidence that platform concentration data (217 ± 164 µg m^{-3}, avg. ± st. dev.) were higher than train cabin data (134 ± 100 µg m^{-3} but 84 ± 36 µg m^{-3} on forced ventilation wagons) for PM10 whilst the PM2.5 levels tended to be more similar (127 ± 136 µg m^{-3} versus 114 ± 98 µg m^{-3}); conversely, higher PM1 levels have been recorded in the train cabins (51 ± 19 µg m^{-3}) than on the platforms (34 ± 15 µg m^{-3}) where they were in the same orders than the subway entrance and exit hall. Platform concentration in our study are in good agreement with those reported for stations with similar design in Barcelona subway system [43]; forced ventilation cabin-to-platform ratios for PM10 and PM2.5 are in the same orders reported for Los Angeles subway system [41]. For UFPs, only a slight, non-significant contrast has been observed between platform and train cabin data, with tendentially higher values for the former dataset ($1.40 \pm 0.43 \times 10^4$ cm^{-3} vs $1.20 \pm 0.37 \times 10^4$ cm^{-3}).

Due to the availability of only a small data set, it was not possible to derive conclusions regarding the conditions that generated the PM concentration peaks. Actually, platform PM10 peaks were mostly, but not systematically, observed at the train arrival in the stations as a consequence of the piston effect; similarly, train cabin PM1 peaks were frequently but not consistently observed at train stops with passengers getting on/off the train. Apparently, the key factors determining the concentration peaks for PM10 were the platform design (namely, its width) and the frequency of the trains; for PM1 the number of people getting on/off and cabin overcrowding.

Statistically significant correlations are observed between PM mass concentration fractions in all the transport modes, with remarkably high values for coarse (PM10–2.5) and fine super micron particles (PM2.5–1) in the car and walk modes (Table 4). Conversely, in all transport modes UFPs are generally not correlated with PM mass, reflecting the presence of different sources and behaviors of large particles, that affect the mass concentrations, and of ultrafine particles that dominate the number concentrations. However, a rather low but statistically significant correlation between UFP and PM1 is found for the car (R = 0.42), supporting the evidence of a different removal efficiency of the car ventilation/filtration system for the sub- and super- micron size fractions. The correlation also suggests that the tailpipe exhaust emissions of the vehicle directly in front, especially in stop-and-go and queuing conditions, are a primary source of in-vehicle sub-micron PM. Such conditions were quite frequent during car mode trips as several crossings regulated by traffic lights are present along the test route.

The results of the cluster analysis point out the peculiar features of the PNSD at the investigated MEs, clearly separating the subway and the car mode from the two open-air modes (bike and walk) where similar particle sizes were found. However, some similarities with the outdoor PNSD were found also in the subway ME, typically in those areas where the air exchange with the outside is higher and the local emissions from train operation have less influence.

Table 4. Correlation coefficients between PM mass concentration and number data for transport modes (italic type: significant correlation at α = 0.05 significance level).

	Car			Bike		
	PM10-2.5	PM2.5-1	PM1	PM10–2.5	PM2.5–1	PM1
PM2.5–1	*0.92*			*0.55*		
PM1	*0.48*	*0.67*		*0.35*	*0.7*	
UFP	0.03	0.01	*0.42*	−0.11	−0.14	−0.04
	Walk			Subway		
	PM10-2.5	PM2.5-1	PM1	PM10–2.5	PM2.5–1	PM1
PM2.5–1	*0.80*			*0.36*		
PM1	*0.07*	*0.26*		*−0.19*	*0.44*	
UFP	−0.08	−0.08	−0.17	0.10	0.18	*−0.21*

3.5. Strength and limitations

The main points of strength of this work are: i) the fact that measurements were taken between the same start/end points for all the travel modes and exactly along the same route for on-road modes; ii) the fact that PM mass concentration for the subway mode were derived through specific number-to-mass conversion factors that account for the different features of the particles emitted in the subway environment. Additionally, to our knowledge, this is the first work investigating commuters’ exposure levels by travel mode in Milan.

However, the work has also some limitations which are mainly related to the features of the dataset. The dataset is actually rather small and limited to the summer period, under fairly good air quality conditions. In winter, when ambient PM and UFP concentrations are much higher, we can expect a greater contrast between the exposure levels for the car and the active modes. Additionally, though providing a direct comparison of the travel modes, our data refer to a rather short urban route that could not be fully representative of the entire commuting routes, as far as concerning the road configuration, traffic intensity, and subway wagon crowding.

As in other studies [9], measurements for the different modes were neither simultaneously nor randomly taken. Though this is not expected to significantly affect our results because all measurements were all taken within a short time window, nevertheless some systematic bias may have been introduced.

Finally, as the data for this study were collected in 2010, an issue can be raised about the road traffic emissions then compared to the current situation. However, from 2010 on, there haven't been substantial changes both in traffic regulation and public transportation services concerning the roads. Therefore, we may assume that the data we present are still relevant as far as traffic flows are concerned. The emission rates may be potentially affected by the renewal of the vehicular fleet which is not particularly fast in Italy (vehicle average age is about 10 years). In any case, the renewal rate hardly followed its "physiological" trend, being limited by the general economical crisis and the lack of monetary incentives for old car scrapping. Nevertheless, a slightly higher share of diesel passenger car equipped with DPF (Diesel Particulate Filter) is nowadays present in the circulating fleet, but this is not expected to significantly affect our data relevancy, at least as far as travel mode comparison is concerned.

4. Conclusion

Exposure levels of PM10, PM2.5, PM1 and UFP characterizing four different transport modes (car, walk, bike and subway) have been measured travelling along a trafficked road in the city centre of Milan. The results of the study indicate that the transport mode significantly affects the exposure levels and point out that the PM mass and number concentrations vary in different ways across the transport modes. The highest values of PM mass concentrations are found in the subway microenvironment, as a consequence of the local PM sources that generate particles with physiochemical features, namely composition and density, different from airborne particles. In contrast, the highest levels of UFP number concentration characterize the two open-air modes (bike and walk), highlighting the influence of direct traffic emissions. On the other hand, the ventilation/filtration system present in the car provides an efficient protection from outdoor particles intrusion into the cabin, especially for the super micron particles for which the ventilation/filtration is more effective. In fact, the lowest average levels of both PM mass and UFP number concentrations are found in this transport mode. However, this latter finding should not be extrapolated beyond the conditions under which it was collected without appropriate caution because it can be influenced by the type of vehicle (and also settings) used during the monitoring campaigns; different results could be obtained when using a small economical car or an older car model.

This work was not intended to assess the integrated exposure and related inhaled dose by transport mode. However, despite some limitations due to the small dataset focused on a rather short route and to the summer period, this work points out that active mode travelling in Milan city centre results in higher exposure levels than the car mode. Walkers and cyclists are expected to experience inhaled quantities and lung deposited dose higher than car passengers due to the combination of higher exposure levels, increased minute ventilation, and longer travel time; their exposure is likely much more higher during wintertime, due to the higher ambient PM and UFP concentration. Thus, the health benefit from active travelling can be partially offset by the higher integrated exposure.

Therefore, with regard to traffic management and air quality policies, the outcomes of the study suggest that, together with road traffic reduction policies, interventions intended to re-design the urban mobility should also opt for the development of dedicated routes for walkers and cyclists in order to limit their exposure to PM and UFP by increasing their distance from road traffic.

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Conflict of interest

All authors declare no conflicts of interest in this paper.

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