Exposure of in-pram babies to airborne particles during morning drop-in and afternoon pick-up of school children

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Graphical abstract

Research highlights

- ▶ PM_{2.5} and PM_{2.5-10} particles dominated morning and afternoon hours, respectively
- ▶ Traffic intersections (TIs) and bus stand emerged as pollution hotspots
- ▶ PNC were by ~65% higher at both TIs and bus stand than at the rest of the route
- ▶ Small differences in PM_{2.5} concentration between babies and adults were noted

Cite this article as: Kumar, P., Rivas, I., Sachdeva, L., 2017. Exposure of in-pram babies to airborne particles during morning drop-in and afternoon pick-up of school children. *Environment Pollution*, doi:10.1016/j.envpol.2017.02.021

Abstract

In-pram babies are more susceptible to air pollution effects, yet studies assessing their exposure are limited. We measured size-resolved particle mass (PMC; 0.25–32 μm) and number (PNC; 0.2–1 μm) concentrations on a 2.7 km route. The instruments were placed inside a baby pram. The route passed through 4 traffic intersections (TIs) and a bus stand. A total of ~87 km road length was covered through 64 trips, made during school drop-in (morning) and pick-up (afternoon) hours. The objectives were to assess PMC and PNC exposure to in-pram babies at different route segments, understand their physicochemical characteristics and exposure differences between in-pram babies and adults carrying them. Over 5-fold variability (14.1–78.2 $\mu\text{g m}^{-3}$) was observed in PMCs. Small-sized particles, including ultrafine particles, were always higher by 66% (PM_{10}), 29% ($\text{PM}_{2.5}$) and 31% (PNC) during the morning than afternoon. Coarse particles ($\text{PM}_{2.5-10}$) showed an opposite trend with 70% higher concentration during afternoon than morning. TIs emerged as pollution hotspots for all the particle types. For example, $\text{PM}_{2.5}$, $\text{PM}_{2.5-10}$ and PNCs during the morning (afternoon) at TIs were 7 (10)%, 19 (10)% and 68 (62)% higher, respectively, compared with the rest of the route. Bus stand was also a section of enhanced exposure to PNC and $\text{PM}_{2.5}$, although not so much for $\text{PM}_{2.5-10}$. EDX analyses revealed Cl, Na and Fe as dominant elements. Road salt might be a source of NaCl due to de-icing during the measurements while Fe contributed by non-exhaust emissions from brake abrasion. The respiratory deposition rates imitated the trend of PMC, with higher doses of coarse and fine particles during the afternoon and morning runs, respectively. Special protection measures during conveyance of in-pram babies, especially at pollution hotspots such as traffic intersections and bus stands, could help to limit their exposure.

Capsule

Exposure assessment of in-pram babies showed much higher fine and ultrafine particles at traffic intersections and bus stands during morning (drop-in) than afternoon (pick-up) hours.

Keywords

Children exposure; Baby pram; Particulate matter; Fine particles; Ultrafine particles

1. Introduction

The negative effects of air pollution exposure on the public health are clearly evident (Heal et al., 2012). A large number of epidemiological studies have associated the exposure to air pollutants to a wide range of respiratory and cardiovascular diseases (among others) and an increase in mortality (Atkinson et al., 2015). For example, Lim et al. (2012) quantified the global burden of disease and attributed 3.2 million premature deaths and 3.1% of global disability-adjusted life years to ambient particulate matter (PM).

Children are considered to be the population group that is most susceptible to environmental

exposures, because of their immature and developing systems, higher inhalation rates and lower body weight with respect to adults (Ashmore and Dimitroulopoulou 2009; Goldman 1995; Peled 2011). Past studies reported the association of increased risk of respiratory diseases and infant mortality with the exposure to air pollutants (Landrigan and Etzel 2014). Some studies have also observed an association of early-life exposure (including the first year of life) to air pollutants with the childhood asthma (Deng et al., 2015; Clark et al., 2010), allergic rhinitis (Deng et al., 2016) and an impaired cognitive development (Basagaña et al., 2016; Sunyer et al., 2016). However, studies showing exposure levels of the infants to particle number (PNC) and mass (PMC) concentrations (Table 1) are still sparse and therefore considered as a part of our investigation in this work.

The urban environments are characterised by appreciable particle number and mass emissions from road traffic and the other anthropogenic sources (Kumar et al., 2016; Kumar et al., 2011; Kumar et al., 2015a). Traditionally, exposure to air pollutants has been assessed through fixed-site air quality monitoring stations measuring at the background or traffic hotspots (Goel and Kumar 2014), but these stations might not be representative of the actual air pollution exposure (Kumar et al., 2015b; Steinle et al., 2013). This is particularly true during commuting since this activity results in an increased exposure when compared to other daily activities and is characterised by a high exposure to time ratio (Buonanno et al., 2013a; Dons et al., 2011; Rivas et al., 2016, 2017), especially in high vehicle density areas (Goel and Kumar 2015a). The reasons for this intense exposure is the proximity to the source (with levels of most air pollutants being particularly high along the busy roads) and the peak concentrations being usually observed during the peak traffic hours (Al-Dabbous and Kumar 2014; Kaur et al., 2005; Morawska et al., 2008; Rivas et al., 2016; Zhu et al., 2002). Particularly high PNC are to be expected on the signalised traffic intersections (TI) due to delay and stop-start conditions of the road vehicles (Goel and Kumar 2015a, b).

Pollutant concentration may vary both horizontally and vertically across the city (Goel and Kumar 2016; Kumar et al., 2008c). Because of their height, children during commuting are relatively closer to the traffic emissions than adults since vehicle tailpipes are usually at a height around 30-60 cm above the ground level. Previous studies observed an exponential decrease with height in concentrations of particle numbers and gaseous pollutants (Goel and Kumar 2016; Kumar et al., 2008b; Kumar et al., 2008c; Vardoulakis et al., 2002). These studies build a generic understanding that much higher concentrations are expected close to the road and thus in the breathing zone of the children.

A few studies have reported exposure assessments at a different height within the first 2 m above the ground that might capture the differences between the children and adults (Table 1). For example, Buzzard et al. (2009) reported that short-term (1.5s, 2.5s) averaged maximum

concentrations of PM at adult head height (~1.65 m at the mouth level) were on average half of those observed at the corresponding height of an infant in a pram (~0.85 m). Likewise, other studies showed PNCs to be about 10% higher at 0.55 m (pram height) than at 1.70 m, which corresponds to an adult face height (Garcia-Algar et al., 2014). Another study by Burtscher and Schüepp (2012) observed 35% higher concentrations of ultrafine particles in a bicycle trailer where a child was located compared with those observed at the breathing height of the bicycle driver. On the contrary, Galea et al. (2014) concluded that children in buggies are exposed to lower PM_{2.5} concentrations than the adults pushing the pram. The morphological and chemical composition of particles is of great importance since these parameters are determinants of the toxicity (De Vizcaya-Ruiz et al., 2006; Decan et al., 2016). Hence, health impairments and their degree might diverge among inhaled materials; although epidemiological studies are still scarce in assessing the effects of individual PM components (Bell 2012; Bilenko et al., 2015; Wyzga and Rohr 2015). Off-line samples can be analysed in the laboratory using microanalysis techniques such as Scanning Electron Microscopy (SEM) to determine morphology and energy dispersive X-ray spectroscopy (EDS) to characterise elemental composition, as used in several environmental studies (Azarmi et al., 2015; Chithra and Shiva Nagendra 2013; Moreno et al., 2015a; Mouzourides et al., 2015; Slezakova et al., 2011).

Besides a limited amount of literature on exposure of babies in the prams, the datasets used in these studies was very limited. Therefore, due to high vulnerability of children (and especially for babies during their first month and years), the assessment of exposure of babies to traffic emissions is of extreme importance and form part of the objectives of this work.

The objectives of this study are to assess exposure to PMC and PNC in various size ranges while travelling in their prams during the morning and afternoon school pick-up/drop-in times. In particular, we aim to (i) determine the concentrations measured inside the baby prams and identify the main pollution hotspots of air pollution, (ii) quantify the PM and UFP exposure and associated particles dose of the babies, (iii) assess the physicochemical characteristics of the particles inhaled by the babies, and (iv) compare the difference in exposures between the babies in the pram and the adult pushing it.

2. Methodology

2.1 Site description

The experiment was carried out on a fixed route starting from the University of Surrey to the Sandfield Primary School in the town centre of Guildford, UK (Figure 1). Most parents carry their babies when dropping-off (morning) and picking-up (afternoon) their school children. The route was selected within the school catchment area so that it is representative of a possible typical route. Only weekdays were considered for monitoring since no school activities take place during the weekend. A total of 64 one-sided runs, giving a total of 32 round trips (University of Surrey – Sandfield Primary School – University of Surrey), were made for

personal exposure monitoring of in-pram babies; of which 17 runs took place in the morning (starting 0800 h, local time) and 15 in the afternoon (starting at 1500 h, local time). The total length of the route was 2.7 km and it took an average of 36.2 ± 2.4 min to walk. The time spent on the route was similar during both the morning (35.6 ± 2.3 min) and the afternoon (36.9 ± 2.3 min) runs.

The route was designed to cross the maximum number of TIs and a central bus stand in order to understand the spatial variability in personal exposure. The selected route passed through the Guildford Bus Station and 5 signalised TIs (Figure 1). The area surrounding the University of Surrey has a low traffic density since none of the main streets passes across the campus. However, local buses frequently cross the campus which may lead to intermittent increments of air pollutants concentrations. The rest of the route goes through a high congested traffic zone. After leaving the University, the route goes on a parallel road to the train railways, passes in front of the Guildford main train station, through the bus station and across some residential area (Figure 1). The Sandfield Primary School is located on a busy street, and it was the final destination of the route.

2.2 Instrumentation and data collection

A GRIMM EDM 107 (GRIMM Technologies Inc.) aerosol spectrometer was used for measuring particles in the $0.25\text{--}32\text{ }\mu\text{m}$ diameter range. The flow of the instrument was controlled by an internal pump and kept at 1.2 l min^{-1} . The instrument reported PM concentrations at a time resolution of 6 sec. It was calibrated just before the measurements and has been successfully deployed in our previous mobile measurements (Azarmi and Kumar 2016; Kumar and Goel 2016).

A P-Trak 8525 (TSI Inc.) was employed for measuring PNC in the $0.2\text{--}1\text{ }\mu\text{m}$ size range. P-Trak is not able to measure particles in the nucleation mode ($<20\text{ nm}$), what might lead to an underestimation of the actual PNC measured in our study (Mishra et al., 2012; Rivas et al., 2017). Actually, previous studies in urban environments (including street canyons) have shown that traffic emissions contribute importantly to the nucleation mode, with a peak below $0.02\text{ }\mu\text{m}$ (Gidhagen et al., 2005; Kumar et al., 2008a; Wehner et al., 2002).

A Dylos DC1700 (Dylos Corp.) particulate matter monitor was used for PNC measurements of small ($0.5\text{--}2.5\text{ }\mu\text{m}$ in aerodynamic diameter) and large ($>2.5\text{ }\mu\text{m}$) particles. It is a particle counter based on light-scattering technology (laser beam operating at 650 nm wavelength) that was initially developed for indoor air quality monitoring (Dylos 2016). The output of the Dylos is particle counts per cubic foot (28.38 lit) of air. The upper concentration limit has been found out by Semple et al. (2013) in chamber experiments to be 65,356 particles per 0.01 cubic foot (equivalent to approximately $1,000\text{ }\mu\text{g m}^{-3}$ for $\text{PM}_{2.5}$), which is an unlikely concentration to be

reached in a typical urban environment, except if being very close to a combustion source such as vehicle tailpipes (Kumar et al., 2009). Previous studies have evaluated the performance of the Dylos, both in indoor and outdoor environments, and indicated that the Dylos is able to accurately determine particle size and operate at a wide range of particle concentration while providing similar results to other available monitors (Northcross et al., 2013). However, Steinle et al. (2015) indicated that Dylos (as a low-cost air pollution monitor) is not aimed to deliver a precision similar to the reference monitors, but to offer an indication of exposure to PM.

The position of both the pram and the instruments was continuously recorded on a second basis (i.e. 1 Hz) using a Global Positioning System (GPS; Garmin Oregon 550). The instruments were placed inside a baby pram to mimic the exposure that a baby would receive while being strolled in a pram. The instruments inside the pram were placed at a height of 0.7 m above the floor. The pram was always pushed in the sidewalk, within a varying distance from the road traffic (between 0.3 and 2 m distance, depending on the sidewalk width).

The Dylos was placed inside the pram for about one-quarter of total runs together with the GRIMM particle spectrometer in order to develop a correlation with their measured PMC in real-world operational conditions, as seen in Supplementary Information, SI, Figure S1; we then used this correlation to make Dylos data comparable to the GRIMM instrument for the rest of the runs at the adult height. From run 17 onwards, the Dylos was carried by the pushing adult in order to quantify the exposure of the adult pushing the baby and compare it with the exposure received by the baby in the pram. The Dylos was placed close to the adult breathing zone between 1.40 and 1.60 m by means of Velcro straps with the inlet looking at the front. A protective cover was placed over the buttons to prevent accidental switch-off of the instrument.

A total of 32 round mobile runs resulted in a 6 sec averaged 22594 data points (37.7h) for GRIMM, 1 sec averaged 129583 data points (36.0h) for P-Trak, and 1 min averaged 22594 data points (10.6h) for the Dylos in the adult position.

2.4 SEM and EDS analysis

Total PM mass was collected on PTFE filters with a diameter of 47mm and a nominal thickness of $\approx 1000 \mu\text{g cm}^{-2}$ using GRIMM 1.107. A total of 3 samples were collected, all of them corresponding to the same route but different days of collection as described in Table S1. A blank filter was also included in the analysis for setting up a reference case.

After carbon coating of the sample surface, particles from all samples were characterised using a JEOL SEM (model JSM-7100F, Japan) with a spatial resolution (depending on the sample) of 1.2 nm at 30 kV and 3.0 nm at 1kV. The JEOL SEM was equipped with EDS, thus being able of obtaining information on morphology and elemental composition (semi-quantitative analysis) of the particles collected on the filters. The analyses were carried out at the

MicroStructural Studies Unit of the University of Surrey (UK). Samples were scanned with a high-energy beam (5-15 kV) of electrons in a raster pattern.

2.5 Exposure assessment

The respiratory deposition dose (RDD) is the product of PMC, deposition fraction (DF) and ventilation rate (VR). The PMC depends only on the outside environment, the DF on particle characteristics (size in particular) and, finally, the VR depends on the age, sex and the activity a subject is performing. The DF varies according to the particle diameter and hence is usually not directly proportional to the mass concentration. The RDD is estimated according to Eq. (1), which is adapted from ICRP (1994) and has been used extensively in the literature (Azarmi and Kumar 2016; Goel and Kumar 2015a).

$$\text{RDD of PM (fractions)} = \text{VR}_j \times \text{DF}_i \times \text{PM}_i \quad (1)$$

Where VR_j is the ventilation rate for the j^{th} individual; DF_i and PM_i are the DF and PMC for each of the i^{th} PM fraction, respectively. DFs were calculated with the mass median diameter (d_p) of PM in various size ranges using the Eqs. (2) and (3) provided by (Hinds 1999):

$$\text{DF} = \text{IF} \left(0.058 + \frac{0.911}{1 + \exp(4.77 + 1.485 \ln d_p)} + \frac{0.943}{1 + \exp(0.508 - 2.58 \ln d_p)} \right) \quad (2)$$

Where IF is the inhalable fraction which is calculated using Eq. (3):

$$\text{IF} = 1 - 0.5 \left(1 - \frac{1}{1 + 0.00076 d_p^{2.8}} \right) \quad (3)$$

3. Results and discussions

3.1 In-pram PMC during mobile measurements

Figure 2 shows the different fractions of PMC during the morning and afternoon runs. A large variability of concentrations was observed between the runs, showing differences up to a factor of 7 between the runs averaged concentrations (Figures 2a-b, Table S2). Run averaged PM_{10} ranged from 14.1 to 78.2 $\mu\text{g m}^{-3}$ during morning runs compared with 18.3 to 120 $\mu\text{g m}^{-3}$ during afternoon runs (Figures 2a-b, Table S2).

The t -test was performed on the data to assess statistically significant differences. A run-wise average of on-road afternoon PM_{10} concentrations ($44.0 \pm 26.3 \mu\text{g m}^{-3}$) was 16% higher than during the morning ($37.8 \pm 16.8 \mu\text{g m}^{-3}$; significant p -value < 0.001 ; Figure 2). This difference was in the opposite direction and much more marked for $\text{PM}_{2.5}$ and PM_1 , having 31 and 47% higher concentrations during the morning ($\text{PM}_{2.5} = 21.5 \pm 14.7 \mu\text{g m}^{-3}$, $\text{PM}_1 = 15.4 \pm 15.7 \mu\text{g m}^{-3}$, p -value < 0.001 in both cases) than in the afternoon runs ($\text{PM}_{2.5} = 16.4 \pm 11.8 \mu\text{g m}^{-3}$; $\text{PM}_1 = 10.5 \pm 10.7 \mu\text{g m}^{-3}$; Figure 2). This results in the $\text{PM}_{2.5-10}$ being significantly higher (70%) in afternoons ($27.6 \pm 17.6 \mu\text{g m}^{-3}$) compared with morning runs ($16.3 \pm 8.5 \mu\text{g m}^{-3}$; p -value < 0.001). Coarse particles are mainly primary particles generated by mechanical processes (Heal et al., 2012) and vehicles can importantly contribute to this fraction by the resuspension of road dust and tyre and brake wear emissions (Amato et al., 2009a; Kumar et al., 2013). On the other side, exhaust emissions contribute more importantly to the fine fraction (Viana et al.,

2008). The lower concentration of coarse particles during the morning runs might be due to the moisture content of the pavement during early morning (owing to the overnight condensation of water due to cold temperatures), which drops the mobility and resuspension of road dust (Amato et al., 2012; Omstedt et al., 2005). Although having a higher traffic intensity during the morning, re-suspension was kept to a minimum owing to the settling of particles because of the wetness of the pavement from the overnight dew. Therefore, higher re-suspension of coarser particles was observed during the afternoon when drier road conditions persisted. However, fresh exhaust emissions are responsible for the higher PMC in the fine fraction during the morning runs. This resulted in about 10% higher total PMCs during the afternoon runs. In fact, this also explains the much higher proportion of the coarse fraction to the total PMC during the afternoon (63%) than during the morning (43%) runs.

A handful of literature is available on exposure to in-pram babies (Table 1). One study on this topic is of Galea et al. (2014) who measured PM_{2.5} measurements in the city of Edinburgh (UK). They performed 6 runs in each of their three selected routes obtaining a range of geometric mean (GM) between 5.9 and 46.6 $\mu\text{g m}^{-3}$, which is similar to the range between 4.8 and 63.5 $\mu\text{g m}^{-3}$ of GM that we obtained.

3.2 Effect of pollution hotspots on enhanced PMC exposure to babies

In order to understand the spatial variability of coarse and fine particles at different parts of the route, Figure 3 shows the spatial variation of PM_{2.5-10} (corresponding to the afternoon run #12 up) and PM_{2.5} (morning run #25 down). Run #12 up and #25 down were selected because their corresponding average PMC (PM_{2.5-10} in #12 up as 21.8 $\mu\text{g m}^{-3}$ and PM_{2.5} in run #25 down as 19.3 $\mu\text{g m}^{-3}$; Table S2) that were closest to the overall average concentrations (21.6 and 19.1 $\mu\text{g m}^{-3}$, respectively). High concentrations were observed at different points of the route, owing to traffic congestion and the surrounding built-up environment. However, high concentrations were consistently observed next to TIs (Figure 3) which has also been reported in previous studies involving pedestrians (Kumar and Goel 2016; Moreno et al., 2015b).

Figure 4 shows the concentrations at different sections of the route: TIs, bus stand and rest of the route (i.e., excluding TIs and bus stand). During the morning runs, the average concentration of PM_{2.5-10} and PM_{2.5} at TIs (18.8 ± 12.2 and 22.5 ± 15.9 $\mu\text{g m}^{-3}$) were 19 and 7% significantly higher than at the rest of the route (p -value < 0.001 from the t -test in both cases), which had corresponding values as PM_{2.5} (and PM_{2.5-10}) as 21.0 ± 14.3 (15.8 ± 7.9) $\mu\text{g m}^{-3}$, respectively (Table S3). In the afternoons at TIs, PM_{2.5-10} (30.1 ± 17.2 $\mu\text{g m}^{-3}$) and PM_{2.5} (17.5 ± 12.3 $\mu\text{g m}^{-3}$) were both by about 10% higher than the corresponding values (27.5 ± 18.1 and 16.0 ± 11.8 $\mu\text{g m}^{-3}$; p -values for PM_{2.5} and PM_{2.5-10} were < 0.001 and 0.03, respectively) at the rest of the route. Therefore, concentrations of both coarse and fine particles were slightly higher at the TIs

compared with the rest of the route (Figure 4). These higher concentrations can be explained due to abrupt changes in driving conditions (e.g., braking, acceleration/deceleration), temporary accumulation of vehicles (Kim et al., 2013; Kumar and Goel 2016) and an increase in fuel consumption due to the acceleration of vehicles (Goel and Kumar 2015a).

Taking a deeper look into PMCs around the TIs, we can observe the highest average concentrations at TI₁ and TI₂ (boxplots in Figure S2, values in Table S3). TI₁ is surrounded by buildings that greatly limit the dispersion of pollutants (Ai and Mak 2015; Weber et al., 2006) while TI₂ is the busiest intersection in the route but located in a more open area that favours the dispersion. This explains why TI₁ has 11% higher concentrations for PM_{2.5} than the rest of TIs and showing usually higher concentrations than TI₂. Generally, TIs also followed the overall trend in which respective concentrations of PM_{2.5} are relatively higher during the morning than the afternoon but an opposite trend was seen for PM_{2.5-10} for the similar reasons discussed in Section 3.1.

At the bus stand, coarse particles were by about 5% ($15.0 \pm 9.0 \mu\text{g m}^{-3}$) and 25% lower than the rest of the route during the morning (no significant differences, p -value = 0.36) and afternoon (significant differences, p -value < 0.001) hours, respectively. On the other hand, fine particles were by about 12% ($23.5 \pm 13.7 \mu\text{g m}^{-3}$) and 21% significantly higher than the rest of the route during the morning (p -value = 0.027) and afternoon (p -value = 0.013) hours, respectively. The higher concentrations of *fine* particles can be explained from exhaust emissions by the running engines of idling buses when passengers get on and off the buses (Figure 4a). Conversely, these stationary idling buses limit the re-suspension of coarse particles explaining their lower concentrations at the bus stands (Figure 4b).

3.3 In-pram PNC during mobile measurements

Figure 5 shows the average PNC for the morning and afternoon trips. The run-wise average PNC was $10155 \pm 6030 \text{ cm}^{-3}$ and $7739 \pm 5004 \text{ cm}^{-3}$ during the morning and afternoon runs, respectively (Figure 5a–b). The average PNC was 31% significantly higher during the morning than afternoon runs (p -value < 0.001), similar to what was observed for PM_{2.5}. These results were the expected since the traffic emissions are an important source of ultrafine particles (Kumar et al., 2014; Morawska et al., 2008) and the highest traffic intensities took place during the morning runs. The average PNCs inside the baby pram were much lower than those reported by roadside measurements in different cities of the UK (Agus et al., 2007; Kumar et al., 2014; Longley et al., 2003; Reche et al., 2011; von Bismarck-Osten et al., 2013), although these studies included smaller particles in their measured PNCs. Since traffic emissions substantially contribute to the nucleation mode particles, especially below 20 nm (Gidhagen et al., 2005; Kumar et al., 2008a; Wehner et al., 2002), an underestimation from the P-Trak is expected because of its detection limit being 20 nm. Garcia-Algar et al. (2015; Table 1) also measured PNC concentrations in the same range (with P-Trak) in different roadside

routes in a baby pram in the city of Barcelona, obtaining an average PNC of $48198 \pm 25296 \text{ cm}^{-3}$. Their value is about 5-times higher than the average PNCs we measured in Guildford. This difference can be explained by the higher background PNC found in Barcelona than in other UK cities which are busier than Guildford (Kumar et al., 2014) and also that their routes were entirely monitored in street canyons characterised by heavy traffic.

Following the methodology used in Section 3.2, we plotted the run (#24 down) that has the trip average PNC closest to the overall average of all runs for assessing the spatial variability (Figure 6). A large variability in average concentrations was noted, showing up to a factor of 8 during the morning ($2965\text{--}23014 \text{ cm}^{-3}$) and a factor of 14 during the afternoon ($1819\text{--}25079 \text{ cm}^{-3}$). PNC at the route section within the University campus were low because of low traffic volume. Thereafter the concentrations started to increase to high levels when approaching the city centre. PNC was found to be consistently higher near the traffic intersection and bus stand compared with the rest of the route (Figure 6d). For example, PNC at TIs during the morning ($14908 \pm 8982 \text{ cm}^{-3}$) and afternoon ($11031 \pm 6683 \text{ cm}^{-3}$) runs were 68% and 62% significantly higher than the corresponding values at the rest of the route ($8890 \pm 5682 \text{ cm}^{-3}$ and $6813 \pm 4850 \text{ cm}^{-3}$; p -value < 0.001 in both cases; Table 2). As previously observed for $\text{PM}_{2.5}$ in Section 3.2 (Figure S3), TI_1 showed the highest concentration among the rest of TIs (around 50% higher) owing to the higher vehicle density in this section of the route and the hindered dispersion by the surrounding buildings. Regarding the influence of the bus stand, PNCs were also much higher in this route section. In line with the $\text{PM}_{2.5}$ (Section 3.2), PNC during the morning ($14396 \pm 9690 \text{ cm}^{-3}$) and the afternoon ($11534 \pm 6809 \text{ cm}^{-3}$) were 62 and 69% significantly higher at the bus stand than at the rest of the route (p -value < 0.001 in both cases), respectively, maintain their reputation as zones of high pollution exposure (Goel and Kumar 2014, 2015b).

3.4 Comparison of babies and adult exposure

We measured $\text{PM}_{2.5}$ concentrations at adult heights to compare with those measured simultaneously inside the baby pram (Figure S4). A huge variability in mean concentration was noted during the morning ($9.6\text{--}36.3 \mu\text{g m}^{-3}$) and afternoon ($9.1\text{--}19.6 \mu\text{g m}^{-3}$) runs for babies. The corresponding range was comparatively smaller for adults during the morning ($9.6\text{--}46.1 \mu\text{g m}^{-3}$) and afternoon runs ($8.3\text{--}16.7 \mu\text{g m}^{-3}$). The mean concentration for overall runs was found out to be nearly identical for babies ($16.4 \pm 9.3 \mu\text{g m}^{-3}$) and adults ($16.9 \pm 10.6 \mu\text{g m}^{-3}$, p -value = 0.102). However, $\text{PM}_{2.5}$ concentrations for babies were about 5% significantly lower than adults during the morning (p -value = 0.016) runs as opposed to 10% higher for babies during the afternoon runs (p -value < 0.001). Although a different instrument was used for adults and babies, the correlation between them was fairly good ($R^2 = 0.94$; Figure S1). We can only compare these results to another study assessing $\text{PM}_{2.5}$ concentrations in a baby pram (Galea et al., 2014). Their results indicate that adults were exposed to higher concentrations than babies. However, their monitoring was limited to only 6 days; they obtained the opposite result (i.e.,

babies exposed to higher concentrations) on one of their six days of monitoring. More long-term studies could further help to establish an exposure gradient between the in-pram babies and adults.

Further inspection of the data for the different parts of the route (Figures 9c-d) showed the mean PM_{2.5} concentration for babies was 5.0 (significant, p -value <0.045), 6.0 (not significantly different, p -value <0.290) and 5.7% (not significantly different, p -value <0.515) lower than adults during the morning hours for the rest of the route, traffic intersection and bus stand sections, respectively. For example, average PM_{2.5} during morning hours were $17.2 \pm 9.8 \mu\text{g m}^{-3}$ (rest of route), $17.6 \pm 8.7 \mu\text{g m}^{-3}$ (traffic intersection) and $19.4 \pm 16.5 \mu\text{g m}^{-3}$ (bus stand) for babies compared with $18.1 \pm 11.4 \mu\text{g m}^{-3}$ (rest of the route), $18.6 \pm 12.3 \mu\text{g m}^{-3}$ (traffic intersection) and $20.5 \pm 10.1 \mu\text{g m}^{-3}$ (bus stand) for adults. The trend changed during the afternoon hours when the mean PM_{2.5} concentrations for babies were 12.5 (significant, p -value <0.001), 1 (not significantly different, p -value <0.724) and 7.7% (not significantly different, p -value <0.508) higher during the rest of the route ($13.2 \pm 5.6 \mu\text{g m}^{-3}$), traffic intersections ($14.0 \pm 5.5 \mu\text{g m}^{-3}$) and bus stand ($14.0 \pm 4.7 \mu\text{g m}^{-3}$), respectively, compared with the adults experiencing $11.7 \pm 3.8 \mu\text{g m}^{-3}$, $13.8 \pm 3.1 \mu\text{g m}^{-3}$ and $13.0 \pm 3.2 \mu\text{g m}^{-3}$ during the rest of the route, traffic intersections and bus stand, respectively. While our results indicate relatively higher exposure to PM_{2.5} to babies during the afternoon runs and lower during the morning hours, past studies also show similar kind of mixed results. For example, Galea et al. (2014) compared the PM_{2.5} concentrations inside a baby pram and at the adult height at 3 different routes over the 6 days of measurements. They generally observed higher exposures of the adults for most days when adult to baby ratios varied between 1.10 and 1.76. They observed an exception on one day when an opposite trend was seen, with an adult to baby ratios being between 0.84 and 0.87; no particular reason for this reversed trend was provided.

Exploring the differences between babies and adults at the different TIs (Table S4) showed no consistent results; sometimes higher exposures for babies than for adults and vice-versa, with most of the TI showing a difference <5% (and always no statistically significant differences). For example, adult to baby ratios at the TIs ranged from 0.97–1.18 during the morning, which changed to 0.88–1.14 during the afternoon. This is expected given that the first few meter height of the TIs is highly affected by the traffic-induced turbulence due to stop-start, acceleration/deceleration conditions (Di Sabatino et al., 2003; Kumar et al., 2008a) and so is the case with the varied emissions and their dispersion (Goel and Kumar 2014, 2015a) contributing to the observed trend.

3.5 Physicochemical properties of the particles

The morphology and the chemical composition was characterised using SEM (Figure 7) and EDS techniques (Table S5). These analyses were performed on the bulk mass of total

PM collected on 3 different filter samples (Table S1). The particles show a wide range of morphologies under the SEM, from agglomerates (related to vehicle exhaust emissions), crustal forms (related to mineral components), as well as the presence of some fibres, probably coming from the fabric of the pram (Figure 7). As expected, the EDS analysis of the blank PTFE filters showed the filter composition, being 88.9 wt% fluorine (F) and 10.8 wt% carbon (C); Table S5. Analysis through SEM-EDS are semi-quantitative, and only the relative contribution of each element was quantified for a specific section of the filter. In Samples 1 and 3, the dominant elements are chlorine (Cl), sodium (Na), iron (Fe), and oxygen (O); excluding F and C that are affected by the filter composition. Road salt can be considered an important contributor to PM in form of sodium chloride (NaCl, characterised by a crystal form) since the measurements were carried out in the winter time when salt is commonly used for de-icing or anti-icing of the roads, especially in the early morning. Fe is a typical crustal element, which is usually found in the form of iron oxides (Fe_2O_3). However, its higher proportion with respect to other crustal elements (Si, Al, Mg) suggests a clear contribution from road traffic, being Fe a tracer of non-exhaust emissions from brake abrasion and commonly found in city dust profiles (Amato et al., 2009b). In Sample 3, the EDS analysis also revealed the presence of Cu in a low wt% (Table S5), which is also a tracer for brake wear (Thorpe and Harrison 2008). Sample 3 corresponds to the sample with the highest bulk mass, mainly due to a higher number of runs, and therefore Cu might have become detectable for the EDS.

3.6 Exposure assessment

The RDD provides the net influx of the PM into the respiratory system and is often cited as the most important factor in determining the ability to adversely affecting the human body. The deposition fraction varies according to the particle diameter and therefore is usually not directly proportional to the mass concentration. We estimated the RDD of different PM fractions for male and female babies during morning and afternoon runs in different parts of the route (Figure 8). As stated in Section 2.5, the RDD is a product of mass concentration, DF and VR (Eq. 1). The former two depends only on the outside environment whereas the ventilation rate depends on the age, sex and the activity, a person is performing. The ventilation rate for the male and female babies of age (<1 year) in sedentary condition is taken as 3.18 and 3.00 lit min^{-1} (US EPA 2009). Since the outside environment is same for both male and female babies during the calculation, the ratio of RDD of the male to the female babies will be fixed as 1.06 at any particular location during any period of the day. Therefore, we only discussed the results for female babies in the subsequent text and the RDD for male babies can be obtained by multiplying the above-noted factor for female babies.

The total RDD rates, which is a sum of fine and coarse particles, for female babies were found to vary substantially from run to run (Figure 8), within a range of 1.2–11.6 $\mu\text{g h}^{-1}$ and 3.0–20.4 $\mu\text{g h}^{-1}$ during the morning and afternoon runs, respectively. The separation of the data

suggested showed around 41% lower average RDD for *coarse* particles during morning runs ($2.8 \pm 1.4 \mu\text{g h}^{-1}$) compared with afternoon runs ($4.7 \pm 2.9 \mu\text{g h}^{-1}$; significant, p -value < 0.001). The corresponding difference reduced to 10% for *fine* particles, with $1.1 \pm 0.6 \mu\text{g h}^{-1}$ during morning compared with $1.0 \pm 0.6 \mu\text{g h}^{-1}$ during the afternoon (significant, p -value < 0.001). This effect was also reflected in the fraction of RDD that changed from 72% (morning) to 83% (afternoon) for coarse particles and from 28% (morning) to 17% (afternoon) for fine particles (Figure 8). The RDD rates mimic the trend of PMC. The abundance of fine particles during the morning hours was expected due to higher vehicle density, as discussed in Section 3.1. Moreover, the wetness of road pavement due to overnight dew and the higher relative humidity during the morning ($84.9 \pm 6.5\%$) compared with afternoon ($71.4 \pm 12.5\%$) might have contributed to limit the re-suspension of coarse particles during morning runs, as also reported by previous studies (Kumar and Goel 2016). The studies quantifying the RDD for babies are scarce for direct comparison. We used a study that estimated RDD for walking along the urban roadsides to put our results in perspective. For example, Nyhan et al., (2014) determined the RDD for 8 adult pedestrians in Dublin and obtained an RDD of $27.6 \mu\text{g h}^{-1}$ for $\text{PM}_{2.5}$ and $40.8 \mu\text{g h}^{-1}$ for PM_{10} , which are much higher than the ones we obtained for in-pram babies. Various factors might explain these differences. Firstly, our average concentrations were lower (overall $\text{PM}_{2.5} = 19.1 \mu\text{g m}^{-3}$; $\text{PM}_{10} = 40.7 \mu\text{g m}^{-3}$) than theirs ($\text{PM}_{2.5} = 28.2 \mu\text{g m}^{-3}$; $\text{PM}_{10} = 45.8 \mu\text{g m}^{-3}$). Secondly, a major effect might be due to the different inhalation rates, which depend on age and intensity of physical activity. The pedestrians assessed by Nyhan et al. (2014) were adults undergoing a moderate exercise (walking) while the in-pram babies were considered in resting position. The ventilation rate for a male adult (30-40 years old) is $30.30 \text{ lit min}^{-1}$ compared with $3.08 \text{ lit min}^{-1}$ for a baby in a sedentary condition (sedentary condition; $< 1 \text{ yr}$) (US EPA 2009), giving an order of magnitude difference and explaining the low RDD in our case for in-pram babies.

In order to assess the spatial variability in the exposure doses over the route, we further analysed the data by dividing our route into traffic intersections, bus stand, rest of the route, and overall route average. As expected, the RDD rates were found to be the highest at the TIs (Figure S5). For example, the mean RDD for coarse particles at the TIs during the morning runs ($3.2 \pm 2.1 \mu\text{g h}^{-1}$) were about twice of those during the afternoon runs ($5.1 \pm 2.9 \mu\text{g h}^{-1}$), demonstrating the dominance of coarse particles during the afternoon runs (significant, p -value < 0.001). The similar trend was evident for bus stand during the morning ($2.6 \pm 1.5 \mu\text{g h}^{-1}$) and afternoon ($3.5 \pm 1.8 \mu\text{g h}^{-1}$) runs (significant, p -value < 0.001). However, the mean RDD for fine particles showed a reverse trend, as was seen for the PMCs (Section 3.1), with $1.2 \pm 0.6 \mu\text{g h}^{-1}$ and $1.0 \pm 0.6 \mu\text{g h}^{-1}$ at TIs (significant, p -value < 0.001) and $1.3 \pm 1.0 \mu\text{g h}^{-1}$ and $1.2 \pm 0.9 \mu\text{g h}^{-1}$ at the bus stands (not significantly different, p -value < 0.269) during the morning and afternoon runs, respectively. An important finding is that in-pram babies experience relatively larger doses of fine particles during the morning while the doses are higher for coarse particles during

the afternoon runs. Fine particles show larger health impacts compared to their larger counterparts (Heal et al., 2012) and at the young age children are more susceptible to particulate pollution due to their immature system, higher inhalation rates and lower body weight (Heinrich and Slama 2007), suggesting a clear need for precautionary measures to limit their exposure during their transport along the busy roadsides.

4. Conclusions

Personal exposure of in-pram babies for both PMC and PNC were assessed along with the concurrent measurements of adult exposure to PM_{2.5}. The objectives were to quantify the in-pram babies exposure of airborne particles when the parents drop-in and pick-up their children during morning and afternoon hours. The specific contribution of traffic intersections, bus stand was also studied.

The following conclusions were drawn:

- PM_{2.5}, PM₁ and PNC concentrations were 47, 31 and 31% higher during the morning runs than those during the afternoon hours, reflecting the influence of traffic emissions during the morning peak hours. Conversely, coarse particles were 70% higher in the afternoons, indicating that re-suspension was importantly affected by the wetness of road pavement due to overnight dew in the early mornings. A similar trend was reflected by the fraction composition showing the proportion of coarse particles as 43% and 63% during morning and afternoon runs, respectively.
- TIs were hotspots of enhanced exposure for PM_{2.5-10} (19 and 10% higher during morning and afternoon, respectively), PM_{2.5} (7 and 10% higher) and PNC concentrations (68% and 62% higher) compared with the rest of the route. TI₁ showed the highest PM_{2.5} and PNC concentrations since it was one of the busiest TIs on the route and the pollutant dispersion here may have been limited by the surrounding buildings. The section crossing the bus stand also showed higher concentrations for PM_{2.5} (12 and 21% higher during morning and afternoon, respectively) and for PNC (62 and 69%) compared with the rest of the route due to the fresh emissions of idling buses, but were lower for PM_{2.5-10} (5 and 25% lower during morning and afternoon, respectively).
- A comparative analysis between the babies and adults showed trivial differences in overall average PM_{2.5} concentrations. Our results showed 5% lower concentrations of PM_{2.5} for babies during morning drop-in hours as opposed to 10% higher concentrations during the afternoon pick-up hours compared with adults.
- A wide range of morphologies was observed under SEM, from exhaust agglomerates to mineral crustal forms and cloth fibres. Non-exhaust elements which are traces of brake abrasion were identified by means of EDS as well as mineral components from road dust re-suspension. Salting the roads might be an important source of NaCl.
- The RDD rates imitated the trend of PMC. A considerable variability was observed

between the hourly RDD estimated for female babies, which were between 1.2-11.6 $\mu\text{g h}^{-1}$ and 3.0-20.4 $\mu\text{g h}^{-1}$ during the morning and afternoon runs, respectively.

This study provides hitherto missing knowledge on the exposure of in-pram babies during the morning and afternoon pick-up periods of children from school. The findings clearly suggest much higher concentrations of fine PMC and PNC during the morning peak hours, especially on the traffic intersections and bus stand. While our exposure assessment and SEM/EDS analyses provided information on respiratory doses and various chemical species, further studies assessing the toxicity of particles in various size ranges are recommended to understand their impact on infant children.

5. Acknowledgements

This work has been carried out under the framework of the University Global Partnership Network (UGPN) funded project, NEST-SEAS (Next-Generation Environmental Sensing for Local To Global Scale Health Impact Assessment). The authors thank Zhenchun Yang, Simon Dai and Monirupa Ananya for their help during the data collection.

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Table 1. Review of studies that have assessed children/babies personal exposure of PM and UFP.

City (Country)	Pollutants	Study design	Equipment Used	Author (year)
Edinburgh (UK)	PM _{2.5}	Measurements at adult and buggy heights on 3 different routes; 6 days, three 1-h trips/day	PM _{2.5} : SidePak AM510 (TSI Inc.) with PM _{2.5} impactor	Galea et al., (2014)
Barcelona (Spain)	UFP (20-1000 nm)	Measurements at buggy height (0.55m) and adult pedestrian (1.70 m) on 3 different walking routes of 5 km. 10 days, total measurements for 20h	UFP: P-TRAK 8525 (TSI Inc.)	Garcia-Algar et al., (2014)
Cassino (Italy)	BC, UFP (10-300 nm)	103 children (8-11 yr) once during 48h	UFP: NanoTracer (Philips) BC: MicroAethalometer AE51 (Magee Scientific)	Buonanno et al., (2013b)
Switzerland	UFP (10-700nm)	Child on a bicycle with a child trailer (measurements at different heights) in different routes	UFP: DiSCmini (Matter Aerosol)	Burtscher and Schüepp (2012)
Morgantown (WV, USA)	PM (mass and particle number counts; 1-1000 nm), CO, CO ₂ , NO _x	Measurements at different heights near a roadway	UFP, PM ₁ : Fast response particle spectrometer (Cambustion DS500) CO/CO ₂ : Horiba AIA-200 NO _x : EcoPhysics CLD-822	Buzzard et al., (2009)
Utrecht (The Netherlands)	PM _{2.5} , Soot, NO _x , NO ₂	54 children (10-12 yr) monitored once during 48h	PM _{2.5} : GK2.05 cyclones (BGI Inc., Waltham, MA) NO _x : Ogawa passive samplers (Ogawa and Company USA, Inc.)	Van Roosbroeck et al., (2007)
Amsterdam (The Netherlands)	PM _{2.5} , Soot, NO _x , NO, NO ₂	14 children (9-12 yr) monitored 4-times during 48h within 2 months	PM _{2.5} : GK2.05 cyclones (BGI Inc., Waltham, MA) NO _x : Ogawa passive samplers (Ogawa and Company USA, Inc.)	Van Roosbroeck et al., (2006)

Table 2. Summary of on-route PNC measurements at different TIs during morning and afternoon runs.

Section	Morning				Afternoon			
	Average $\pm$ SD	Median	Min	Max	Average $\pm$ SD	Median	Min	Max
Rest Route	8916 $\pm$ 9689	6100	482	205000	7050 $\pm$ 8574	7420	560	157000
TI all	14908 $\pm$ 8982	12893	3997	34052	11031 $\pm$ 6683	8889	3721	33260
TI ₁	20062 $\pm$ 13282	15851	1762	53546	16455 $\pm$ 12729	11047	1706	50365
TI ₂	12058 $\pm$ 7361	9297	847	30998	9718 $\pm$ 7154	7518	1669	36356
TI ₃	13974 $\pm$ 10574	11448	675	41317	11251 $\pm$ 10425	8770	1677	53499
TI ₄	14223 $\pm$ 14104	8002	1377	54863	10784 $\pm$ 7303	9088	1739	28535
Bus Stand	14396 $\pm$ 9690	11914	2043	49561	11534 $\pm$ 6809	9271	3019	24731

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Figure 1. The map shows the route followed and indicates the location of the origin and destination points along with the bus stand and the traffic intersections crossed.

Figure 2. Concentrations of different PM types during (a) morning, (b) afternoon runs. The whiskers show the 5% and the 95% percentile while the cross sign shows the average value. In figures (a) and (b) each bar represents one-way trip between the origin (University of Surrey, UoS) and destination (Sandfield Primary School, SPS). Please note that each trip show two bars and the first and the second bars represent Up (UoS-SPS) and Down (SPS-UoS) trips, respectively. In figures (c) and (d), boxplots of PM_{2.5-10} and PM_{2.5} concentrations are shown for morning and afternoon runs; overall represent the average of both morning and afternoon runs.

Figure 3. Spatial variability of (a) coarse particle concentration during run #12 up, and (b) fine particles measured at the route during run #25 down. The PMC range for each fraction was split into 5 colour categories by referring to the corresponding legend.

Figure 4. (a) PM concentrations and (b) their fractions during morning and afternoon runs at the traffic intersections, bus stand and the rest of the route.

Figure 5. (a) Run wise concentrations of PNCs during (a) morning, (b) afternoon, (c) the averaged concentrations during morning and evening runs, (d) together with overall PNCs at different parts of the route. The whiskers show the 5% and the 95% percentile whereas the average values are shown by the cross signs.

Figure 6. Spatial variability of ultrafine particles at the route. This GIS maps represent run #24 down, which have average concentrations similar to the overall average of the entire runs.

Figure 7. SEM images of the mass collected on the PTFE filters during the different runs.

Figure 8. Respiratory deposition doses of in-pram babies during (a, b) morning, and (c, d) afternoon up and down runs. Please note that each run presents two bars for up and down measurements of the route, respectively. Each run numbers presents a return trip between the origin and destination points. The runs #24 and #29 did not show data because of the rain during these run periods. Sub-figure (e) and (f) show the RDD of fine and coarse particles, inside the baby pram during the morning and afternoon runs, respectively; overall represents the average of morning and afternoon runs. The whiskers show the 5% and the 95% percentile whereas the average values are shown by the cross signs.