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K.V. Abhijith, Prashant Kumar

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CRedit authorship contribution statement

KV Abhijith: Conceptualization, Methodology, Validation, Formal analysis, Investigation, Data curation, Writing - original draft. **Prashant Kumar:** Conceptualization, Methodology, Investigation, Funding acquisition, Project administration, Resources, Supervision, Writing - review & editing.

Graphical abstract

Quantifying particulate matter reduction and their deposition on the leaves of green infrastructure

KV Abhijith¹, Prashant Kumar^{1,*}

¹Global Centre for Clean Air Research (GARE), Department of Civil and Environmental
Engineering, Faculty of Engineering and Physical Sciences, University of Surrey, Guildford
GU2 7XH, United Kingdom

Abstract

The green infrastructure (GI) is identified as a passive exposure control measure of air pollution. This work examines particulate matter (PM) reduction by a roadside hedge and its deposition on leaves. The objectives of this study are to (i) quantify the relative difference in PM concentration in the presence of GI and at an adjacent clear area; (ii) estimate the total mass and number density of PM deposited on leaves of a hedge; (iii) ascertain variations in PM deposition at adult (1.5m) and child (0.6 m) breathing levels on either side of a hedge; (iv) illustrate the relationship between PM deposition to leaves and ambient PM concentration reductions; and (v) quantify the elemental composition of collected particles of the leaves on different heights and sides of hedge. PM reduction of 2-9% was observed behind hedge compared to a clear area and followed a trend of $\Delta PM_1 > \Delta PM_{10} > \Delta PM_{2.5}$. Counting of particles was found to be an effective method to quantify deposition than weighting methods. Sub-micron particles (PM_1) dominated particle deposition on leaves at all sampling points on both sides of the hedge. PM mass deposition and number concentration to the leaves on traffic-facing side was up to 36% and 58% higher at 0.6m compared with 1.5m

*Corresponding author. Address as above. Email: p.kumar@surrey.ac.uk; Prashant.Kumar@cantab.net (P. Kumar)

height, respectively. Such a difference was absent on the backside of the hedge. The SEM-EDS analysis showed up to 12% higher traffic-originated particles deposited to leaves on the traffic-facing side compared to the backside. The naturally occurring particles dominated in identified particles on leaf samples from all collection points on the hedge. These new evidence expand our understanding of PM reduction of GI in the near-road environment and its variations in particle deposition, depending on height and sides of GI, which could allow a better parameterisation of dispersion-deposition models for GI assessment at micro-scale.

Keywords: Green infrastructure; Particulate matter; PM Deposition; Exposure mitigation; Leaf characteristics

1. Introduction

Air pollution is a major cause of premature death in Europe, responsible for more than 400,000 premature deaths (EEA, 2019; HEI, 2018). In the UK, traffic-generated pollution has been identified as a major contributor to particulate matter (DEFRA, 2017). Considerable research findings have indicated a variety of adverse health impacts for those who spend significant amounts of time near major roads (HEI, 2010). Such impacts are attributed to relatively high concentrations of air pollutants within a few hundred metres of the road (Karner et al., 2010; Kimbrough et al., 2018). Passive control measures such as green infrastructure (GI) and noise barriers are identified as near-term mitigation strategies for reducing air pollution exposure near highways (Abhijith and Kumar, 2019; Baldauf, 2017; Deshmukh et al., 2018; Gallagher et al., 2015; Lee et al., 2018). These methods can be implemented and managed by urban developers and planners as complementary pollution control measures as well as in future regulatory plans.

GI may abate PM exposure primarily by enhancing dispersion and providing deposition and absorption surfaces for different pollutants (Janhall, 2015). Comprehensive reviews have

summarised the potential effectiveness of GI for reducing PM exposure in near-road environments (Abhijith et al., 2017; Gallagher et al., 2015; Janhall, 2015; Barwise and Kumar, 2020).

Field experimental studies have reported greater than 50% reductions in various pollutant concentrations behind GI (Ottosen and Kumar, 2020; Abhijith and Kumar, 2019; Deshmukh et al., 2018). The magnitude of reduction in PM concentration is influenced by meteorological conditions and GI factors such as leaf area index (LAI), height, barrier thickness, and stand density (Abhijith et al., 2017; Al-Dabbous and Kumar, 2014; Ghasemian et al., 2017). Several modelling studies revealed that the dispersion component in pollutant reduction dominates the impact of deposition to GI (Buccolieri et al., 2018; Jeanjean et al., 2016; Xing and Brimblecombe, 2019). Nevertheless, many experimental studies have confirmed the importance of GI in PM removal by quantifying significant PM deposition to leaf surfaces. Uncertainties and differences in PM deposition estimates between modelling and experimental studies may be attributable to differences between simulated (for example, *Picea abies*, $V_d = 0.02 \text{ cm s}^{-1}$; Peters and Eiden, 1992) and measured ($V_d = 0.55 \text{ cm s}^{-1}$; Bunzl et al., 1989) deposition velocities.

Studies that have quantified PM deposition to leaves have followed direct, gravimetric methods and indirect, microscopy imaging techniques. In gravimetric methods, leaves are washed with micro-distilled water and any particles are passed through pre-weighed filters with different pore sizes corresponding to different PM classifications (Chen et al., 2017a; Liu et al., 2018). The subsequent residues on dried filters are then weighed for PM quantification. One drawback of this method is that particles may be retained in leaf waxes and leaf micromorphological structures. Additionally, chloroform is the employed solvent for these particles and it can potentially dissolve some of the particles containing non-polar

molecules (Przybysz et al., 2014; Sgrigna et al., 2015; Song et al., 2015). In microscopy methods, highly magnified images of deposited particles are taken, often using a Scanning Electron Microscope (SEM), and particles in these images are counted via image processing software and techniques (Lin et al., 2018; Weerakkody et al., 2019, 2018a, 2018b). This method of analysis also indicates PM size distribution, as well as providing insights into leaf micromorphology. However, SEM only presents tiny fractions of the total leaf surface area, which means that statistically significant numbers of micrographs must be taken to draw any conclusions from analysis (Ottel et al., 2010; Weerakkody et al., 2017).

There is a need for simultaneous evaluation of these two removal mechanisms through field campaigns to determine the relative contribution of each in total air pollution improvement. Uncertainties between modelling studies and field campaigns in quantifying PM deposition to GI may thereby be reduced. The primary objective of the present study was therefore to quantify the deposition and overall PM reduction by GI in near-road environments. An overall reduction in PM was estimated by comparing ambient concentrations behind and in front of a hedge with those of an adjacent clear area along the same busy road in Guildford, UK. In addition, a representative number of leaves at different heights of the hedge were collected to estimate total PM deposition by SEM microscopy and image analysis. The collection of leaves from both sides of the hedge, at adult (1.5m) and child (0.6 m) breathing levels, enabled us to study variations in deposition within the hedge. Finally, analysis of the elemental composition of particles on filter papers of an air sampler and leaves at different heights and sides of hedge revealed the heavy elements removed by GI from the ambient air. This study combines leaf-sampling and ambient air monitoring approaches in quantifying ambient PM removal by GI in near-road environments and elucidates in the relative contributions of deposition to overall PM reductions. More importantly, the study provides

new insights into mechanisms of GI that influence pollution exposure reduction and lead to improvements in human health.

2. Methodology

2.1 Site description

The field campaign consisted of measuring ambient PM concentration with and without green infrastructure and leaf sampling for PM deposition along Stoke Road in Guildford, Surrey, UK. Car travel is the most common form of transportation, including 72% and 42% of all commuting to work and school, respectively (Al-Dabbous and Kumar, 2014). As a two-lane busy road connecting two major towns (Woking and Guildford, Surrey), approximately 1200 vehicles per hour used Stoke Road. The measurement location included a clear area with no obstruction to the airflow and a long hedgerow fencing a children's play area adjacent to Stoke Road (Figure 1). This well-maintained Beech (*Fagus sylvatica*) hedge lies 36 meters along the road with an average height and thickness of 2.2m and 1.5m, respectively (Figure 1). *Fagus sylvatica* is identified as one of common hedge plant in north-western Europe with a high level of ecosystem service delivery, including high PM and gaseous pollutant capture (Blanusa et al., 2019 and references therein). This native European species has broadly elliptical leaves with deciduous foliage; it has weakly developed a smooth layer of wax on the leaf surface (Cho et al., 2014; Tomaszewski and Zieli, 2014). In addition, it has shown high air pollution tolerance and low generation of ground-level O₃ (Blanusa et al., 2019 and references therein). The leaf area index (LAI) of the hedge was measured using a handheld Ceptometer (AccuPAR LP-80, Meter Environment) multiple times during the field campaign, and the average LAI value was 4.47 m²m⁻².

2.2 Sampling and data collection

In this study, there were two simultaneous monitoring points at both the clear area and the hedge site, as shown by sites (1) and (2) in Figure 1, respectively. The sampling height was maintained at an adult breathing height of 1.7m by mounting all instruments on tripods. At each site, there was one measurement point on the roadside footpath (INF) and one behind the hedge or equidistant from the pavement (BHD). We measured PM (PM_1 , $PM_{2.5}$, and PM_{10}) alongside continuous meteorological monitoring and intermittent traffic counting. Aerosol spectrometers (GRIMM models EDM 107 and 11-C) were used to record PM mass concentrations on 31 different channels at six-second intervals (Abhijith and Kumar, 2019; Azarmi and Kumar, 2016; Rivas et al., 2017; Vesa Yli-Pelkonen, Heikki Setälä et al., 2017). All instrument data points were averaged to one minute for further analysis. Following previous studies, meteorological factors including wind speed, wind direction, humidity and temperature were obtained from the nearest UK weather station, located in Farnborough 10 km from the monitoring site (Abhijith and Kumar, 2019; Al-Dabbous and Kumar, 2014; Goel and Kumar, 2016). A portable weather station (Kestrel 4500) recorded in-situ micrometeorological conditions at a height of 1.7m. During each day of monitoring, the traffic volume was counted for 20 minutes every hour. This work collected high-resolution ambient air pollutant concentration data for five days at each site, starting and finishing at around 08:00 and 18:00h, respectively, and thereby capturing the morning and evening traffic peaks. Since the concentrations were assessed in relative terms before the after the GI and that the distance between the simultaneous monitoring points was within two meters, the effect of background concentrations can be assumed to be the same at both sampling points and thus their modest influence on the measured concentrations. To ensure accuracy in pollutant concentration measurements by the similar sets of instruments, collocated runs were performed as in previous studies (Abhijith and Kumar, 2019; Brantley et al., 2014; Lin et al., 2016). Instruments were kept together for 30 minutes of sampling before and after data

collection on each day. All instruments displayed good agreement in estimating concentration levels and the GRIMM instruments showed an inter-relation in estimates with R^2 values of 0.84, 0.97, and 0.96 for PM_{10} , $PM_{2.5}$ and PM_1 , respectively (SI Figure S1). No correction factors to the data were therefore applied.

2.3 Leaf sampling

To quantify PM deposition to leaves, a total of 40 healthy, undamaged leaf samples were collected and preserved for analysis. To distinguish PM deposition to leaves at the children and adult breathing heights, we harvested 10 random leaves at heights of around 0.6m and 1.5-1.7m from each side of the hedge. Leaves were extracted, touching the only petiole, using latex gloves and scissors that were cleaned after each leaf extraction. The harvested samples were stored in plastic sealable bags lined with aluminium foil for structural stability by avoiding further contamination and limiting contact between leaf blades and other surfaces. Leaves exposed to traffic-generated PM were sampled after 12 dry days in early October, when the deciduous leaves were at maturity and before senescence, following previous studies (Shi et al., 2017; Sæbø et al., 2012). The accumulation of the PM deposition on leaves reached its peak in ~24 days after a rainy day, and there was a negligible increase in the amount of deposited PM on leaves afterwards. This time required to reach the maximum PM deposition on leaves were influenced by the surrounding environment, climatic conditions and plant species (Liu et al., 2013). Previous studies have collected leaves after 10 days (Mori et al., 2018), 14 days (Xu et al., 2017) and 20 days (Liu et al., 2018) without rainfall, which were considered to be substantial dust retention periods before reaching maximum PM accumulation. Other studies have undertaken sampling less than five days after precipitation (Chen et al., 2016; Castanheiro et al., 2016; Wang et al., 2015; Yan et al., 2017). We assumed that leaves are clean immediately post rainfall (Chen et al., 2016) and that our sampling was carried out during a period of PM accumulation. To minimise the

influence of weather, the sampling day was sunny, dry, and without high wind speeds ($>3\text{m/s}$) and details are provided in SI Table S1.

2.4 Quantification of PM deposition

Samples for SEM imaging and elemental analysis were prepared from archived leaves, following preparation methods described by Ottel et al. (2010) and Weerakkody et al. (2017). Four sections, $5.0\text{mm} \times 5.0\text{mm}$ in area, were removed from each leaf blade (i.e. either side of the midrib) for microscopy. Two sections were selected at random for the examination of PM deposition on the adaxial and abaxial surfaces of each leaf sample. Leaf sections were mounted on aluminium stubs using double-sided carbon tape. Leaves were dried at room temperature and stored in desiccators. Micrographs were taken using a Scanning Electron Microscope, (JEOL SEM, model JSM-7100F, Japan) equipped with an energy dispersive X-ray spectrometer, using Backscattered Electrons (BSE) at 15 kV, in high vacuum and with-carbon coating (SI Figure S2). Four randomly selected points on each leaf sample were micrographed in series, with a constant centre of view at two different magnifications: $500\times$ and $1200\times$, targeting coarse ($2.5\text{-}10\mu\text{m}$) and fine ($1\text{-}2.5\mu\text{m}$, and $0.27\text{-}1\mu\text{m}$) PM size ranges, respectively. All micrographs were taken at the same working distances, and with consistent contrast and brightness levels, to ensure uniform image procurement procedures for all samples.

Pathfinder 2.0 X-ray Microanalysis Software (Thermo Fisher) was employed to quantify the PM number density for different fractions ($\text{PM}_{10\text{-}2.5}$, $\text{PM}_{2.5\text{-}1}$, $\text{PM}_{1\text{-}0.27}$) on each micrograph, by using binary BSE micrographs and counting the pixels composing each particle. The software calculated the area and circularity of each identified particle, visualised as white against the black leaf surface. The software was initialised with a setting to identify adjoining particles and remove particles consisting of only one pixel. This image data treatment methodology

enabled the software to capture particles effectively and subtract leaf surfaces. The lowest particle diameter detected was $0.27\mu\text{m}$, which comprised more than two pixels in the chosen image conditions and at the highest magnification of $1200\times$ (resulting in a micrograph with an area of $8223.61\mu\text{m}^2$ at a resolution of 1024×768 pixels) (Dappe et al., 2018; Shi et al., 2017). A total of 640 micrographs were analysed to estimate mean PM densities. The mean PM densities (per 1mm^2) on both abaxial and adaxial surfaces were calculated by taking the mean PM densities of all corresponding micrographs. These values were then combined to determine an overall PM density for each leaf. A final mean PM density was estimated by combining values from 10 leaves from each location (height and side) of the hedge. Following this methodology, mean PM diameters for three size fractions ($\text{PM}_{10-2.5}$, $\text{PM}_{2.5-1}$, and PM_1) were also estimated. The total amount of deposited PM per unit surface area of hedge during the 12 days of sampling was calculated from the mean PM density (N_i , $\# \text{mm}^{-2}$), mean PM diameter (D_i , μm), density of the particle (ρ , g cm^{-3}) and the leaf area index (LAI) of the hedge.

$$\text{PM deposition : } PM_i(\mu\text{g cm}^{-2}) = N_i \times 10^{-4} \times \frac{4}{3}\pi \frac{D_i^3}{8} \times \rho \times LAI \quad (1)$$

PM_i is PM ($\text{PM}_{10-2.5}$, $\text{PM}_{2.5}$ or PM_1) deposition ($\mu\text{g cm}^{-2}$). LAI is the total leaf surface area per unit ground area (m^2m^{-2}). The density of particles was taken as 2 g cm^{-3} (Lin et al., 2018; Zhang and He, 2014).

2.5 Comparisons of deposition and total pollution reduction by the hedge

Ambient measurements provided instantaneous concentration change data for both scenarios (i.e. in the absence and presence of the hedge). This time series was converted to a daily average for comparison between the two monitoring locations as well as with leaf deposition. Total PM deposition, quantified by image analysis, was applied as part of a widely used equation (2) for calculating deposition velocity v_d (Janhall, 2015).

$$\text{Deposited amount} = LAI v_d Ct \text{ (g m}^{-2}\text{)} \text{ or } LAD v_d Ct \text{ (g m}^{-3}\text{)} \quad (2)$$

Where *LAI* is leaf area index, *LAD* is leaf area density, v_d is deposition velocity, *C* is average PM concentration at the hedge site, and *t* is the time available for particle deposition to leaves (12 days). The relationship between PM deposition and ambient PM concentration was assessed by comparing time series plots of average ambient PM concentration (with and without the presence of the hedge, from field measurements) with average PM deposition according to equation (3).

$$PM \text{ deposition} = LAD v_d C \text{ (gm}^{-3}\text{s}^{-1}\text{)} \quad (3)$$

Where *LAD* is leaf area density, *C* is the ambient concentration of air pollutants, and v_d is deposition velocity, calculated from leaf deposition quantification as described by equation (2).

2.6 Elemental analysis of particles collected on leaves

The elemental composition of bulk particles collected on leaf samples was determined via SEM-EDS analysis. Particles deposited on leaf samples of the hedge at a breathing height of children (0.6m) and adults (1.5-1.7m) were collected for characterising their chemical composition. Leaf samples were prepared for the quantification of PM elemental composition using a Scanning Electron Microscope (JEOL SEM, model JSM-7100F, Japan) equipped with an energy dispersive X-ray spectrometer at the Micro-Structural Studies Unit of the University of Surrey, UK. The SEM has a spatial resolution of 1.2nm at 30 kV and 3.0nm at 1 kV. The SEM was employed at an acceleration voltage of 10kv, with a working distance of 10mm under vacuum conditions. Automated EDS analyses were performed on contrasting Backscattered Electron (BSE) images with bright white collected particles against the black background of the filter paper using Pathfinder 2.0 software (Thermo-Fisher), following procedures described by Abhijith and Kumar (2019). A total of 2 micrographs were

taken from each sample with an estimated 3000 individual particles at each leaf collection point at the children and adult breathing heights on both sides of the hedge.

3. Results and discussion

3.1 Differences in PM concentrations between GI scenarios and adjacent clear areas

A summary of PM concentration data is provided in terms of descriptive statistics with percentage differences of the mean (SI Table S2) and as box plots (Figure 2). In comparing PM concentrations between monitoring points (BHD and INF) at both sites, higher PM concentration reductions were observed in the presence of GI, whereas the clear area exhibited negligible or no reductions. The order of significance in reductions for PM at the GI site followed $\Delta PM_1 > \Delta PM_{10} > \Delta PM_{2.5}$ as reported earlier in Abhijith and Kumar (2019). The monitoring points (BHD and INF) at the GI site presented greater variation in PM concentrations (maximum and minimum PM values in SI Table S2) than the monitoring points at the clear area. Moreover, the PM build-up in front of the hedge resulted in higher maximum PM values at this monitoring point (i.e. INF at GI site, unlike INF at the clear area), supporting previous results (Abhijith and Kumar, 2019). GI was found to shift the mean PM values above the median close to the upper quartiles, as seen in the boxplot (Figure 2), indicating a high skewness at GI site that is usually absent in case of clear area (Deshmukh et al., 2018). Percentage differences in PM_1 (9%) and PM_{10} (7%) at the GI site were significantly larger than those at the clear area (<3%), as shown by Figure 3. In agreement with previous studies, $PM_{2.5}$ displayed the least percentage differences at both sites, indicating a minimal influence of GI on $PM_{2.5}$ removal (Abhijith and Kumar, 2019; Brantley et al., 2014; Viippola et al., 2018). Coarse PM ($PM_{10-2.5}$) presented the greatest concentration reduction (21%) at the GI site, and a 14% reduction at the clear area may be solely attributed to dilution between monitoring points. The percentage reductions in PM

reported by this study (2-9%; autumn season) were lower than those found by a previous study (15-25%) conducted during summer and carried out at the same site (Abhijith and Kumar, 2019). This observation offers preliminary indications of a steady increase in PM concentrations behind roadside hedges from leaf maturity onwards, and a reversal of the sudden step in PM concentrations as reported by Ottosen and Kumar (2020). This structural change was reflected in LAI of deciduous hedge which reduced from $6.64 \text{ m}^2\text{m}^{-2}$ in summer to $4.47 \text{ m}^2\text{m}^{-2}$ in autumn (Abhijith and Kumar, 2019) displaying its impact on PM concentraion during seasonal variation.

Further insights are provided by visualising hourly averaged relative PM concentration ($\text{PM}_{\text{BHD}}/\text{PM}_{\text{INF}}$) together with boxplots for both monitoring sites, as shown in Figure 4. Scatter plots of relative PM concentration were plotted, and a smooth line was fitted by the *loess* method in R (R Core Team, 2018). Smooth lines in hourly average time series demonstrated an improvement in air quality behind GI, with a lower relative concentration (<1) for the GI site (green line) than for the clear area (red line). This improvement in relative concentration was greatest for PM_{10} , followed by $\text{PM}_{2.5}$, and least for PM_1 . A similar trend was reported by Ottosen and Kumar (2020). As specified in absolute PM concentration values earlier (Figure 2), relative PM concentrations at the GI site had larger variations, which was visible in all boxplots in Figure 4. All relative PM concentration values at the GI site were below 1 and below the smooth line of the clear area (red line), providing evidence of the PM removal potential of GI (Figure 4).

3.2 Deposited flux at leaf-level and PM reductions due to hedge

PM densities on both abaxial and adaxial leaf surfaces from different leaf sampling locations are presented in Figure 5, and summary statistics of PM densities are provided in SI Table S3. The fine (PM_{10}) size fraction accounted for the highest PM density across all leaf sampling locations and it was approximately 10-times higher than the lowest PM density

(coarse PM; $PM_{10-2.5}$), comparable to a 9-fold difference reported by Weerakkody et al. (2017). In agreement with findings from previous studies, the order of PM density was $PM_1 > PM_{2.5-1} > PM_{10-2.5}$, accounting for 66%, 29% and 5% of total deposited particles, respectively, as shown in SI Figure S3b (Ottel et al., 2010; Shao et al., 2018; Shi et al., 2017; Song et al., 2015; Wang et al., 2015; Weerakkody et al., 2017). This demonstrates that leaf surfaces are effective in removing fine particles from the ambient air, and that particle counting methods are more suitable than weighting methods for quantifying deposition of this harmful pollutant to leaves (Ottel et al., 2010; Shi et al., 2017). Interestingly, PM densities across all size ranges were lowest on leaves from a height of 1.5m and on the traffic-facing front side of the hedge ($F_{1.5}$), when compared with the other three-leaf sampling locations. The density of coarse particles ($PM_{10-2.5}$) on leaves at the front side of the hedge (at both 1.5m and 0.6m heights, $F_{1.5}$, $F_{0.5}$) was lower than on leaves at the backside of the hedge ($B_{0.6}$ and $B_{1.5}$; Figures 5a and 5e). Fine PM density on leaves collected at 0.6m height on the traffic-facing side ($F_{0.6}$) was higher than on leaves collected at 1.5m ($F_{1.5}$) on the same side. Sampling points on the backside of the hedge ($B_{0.6}$ and $B_{1.5}$) reported higher or similar PM densities. In examining and comparing adaxial and abaxial leaf surfaces, larger PM densities were found on adaxial surfaces, as represented by Figures 5a-d. This trend, which has also been identified in previous studies, could be due to the favourable orientation of adaxial surfaces for gravitational settlement and the comparatively complex micromorphology (e.g. presence of hairs) of adaxial surfaces (Ottel et al., 2010; Shao et al., 2018; Shi et al., 2017; Song et al., 2015; Wang et al., 2015; Weerakkody et al., 2017). Variation in PM density on abaxial surfaces was less significant than on adaxial surfaces across leaf sampling locations. As seen in SI Figure S3a, 18-25% of all particles were accumulated on abaxial surfaces, which is consistent with reported ranges of 24% and 17% by Shi et al. (2017b) and Wang et al. (2006), respectively. In conclusion, we found higher particle deposition of most size ranges on leaves

at the back of hedge than on the traffic-facing front side of the hedge, and adaxial leaf surfaces captured up to three times more particles than abaxial surfaces. This could be arising from higher traffic-induced turbulence along the traffic-facing side, leading to less deposition. Conversely, the relatively less turbulent conditions at the backside of the hedge may have allowed increased PM deposition to leaves. Moreover, visual inspection of hedge during the field campaign showed smaller and less healthy leaves at traffic facing side compared to large and healthier leaves on the back of the hedge, as showed in SI Figure S4. Exposure to fresh traffic fumes may be the reason for visual differences in the health of leaves from both sides. This could presumably affect the PM retention on leaves. Further studies investigating air pollution tolerance index and air pollution stress on GI could provide their impact on air pollution reduction and PM removal by GI. These results also suggest that it would be beneficial to plant high air pollution tolerant variants close to traffic facing side while designing GI barrier with multiple rows of various species.

The average mass of deposited particles per unit area of the hedge (across sampling points) was estimated by incorporating the mean aerodynamic diameter of particles (SI Table S4), mean PM density (SI Table S3) and LAI of the hedge, and was tabulated in SI Table S5. The mean deposited mass followed an opposite trend to that of PM density ($PM_1 < PM_{2.5} < PM_{2.5-10}$), as expected following previous studies (Mori et al., 2018; Sæbø et al., 2012; Song et al., 2015). The highest mass deposited was of the coarse size range ($PM_{2.5-10}$) and the least was fine (PM_1), accounting for 73% and 3% of mean particle deposition to the hedge, respectively (SI Figure S3c). The deposited amount of $PM_{2.5-10}$, $PM_{1-2.5}$ and PM_1 ranged from 13-17 $\mu g\ cm^{-2}$, 4-5 $\mu g\ cm^{-2}$ and 0.5-0.6 $\mu g\ cm^{-2}$, respectively (Figure 6). Our finding of $20 \pm 2\ \mu g\ cm^{-2}$ mean PM deposition to the *Fagus sylvatica* hedge is generally consistent with the $\sim 17 \pm 3\ \mu g\ cm^{-2}$ described by Sæbø et al. (2012), who collected leaves during the same seasonal period in Norway and Poland. Like the PM density, PM deposition (both fine and coarse) on leaves

at 1.5m height on the traffic-facing front side of the hedge reported least than leaves from remaining sampling locations as observed in Figure 6. Coarse ($PM_{10-2.5}$) particle deposition on the traffic-side was lower than that on the backside of the hedge (Figure 6). The highest PM deposition was observed at 0.6m height on the front, traffic-facing side of the hedge, and mean PM_1 deposition to leaves displayed less variation between sampling points (Figure 6).

Table 1 provides further insight into the impacts of different sampling heights and sides of the hedge. Significant differences in PM deposition and, particularly, PM density were observed between leaf sampling heights for the traffic-facing front side of the hedge, whereas no such trend was observed for the backside of the hedge (Table 1). Leaves closer to the ground (0.6m) were closer to the traffic emission source and subject to less traffic-generated turbulence than leaves at 1.5m, resulting in greater PM deposition. In addition, the barrier effect of the hedge may lead to higher concentrations at ground level than at the upper canopy of the hedge (2.2m), further contributing to increased PM deposition at the $F_{0.6}$ sampling point (Table 1). A similar increase in the number of deposited particles with decreasing sampling height was reported by Mori et al. (2018). Further studies are required to provide a clearer understanding of the complex process of deposition to leaves at different heights of the traffic-facing side of roadside hedges. In contrast, the back of the hedge may be subject to fewer airflow fluctuations on the wake-side of traffic-generated turbulence, thus presenting uniform deposition and PM density on leaves across different sampling heights, as shown in Table 1. Finally, in comparing PM density and deposition to leaves from the same sampling height on both sides, low or very low variations were observed between sampling points at 0.6m height except for coarse PM deposition. As earlier illustrated, the traffic-induced turbulence and upward flow resulting from the barrier effect of the hedge may lead to significantly lower PM densities and PM deposition in the front side of the hedge than on the backside (Table 1).

3.3 Influence of leaf deposition on ambient PM concentration behind GI

Our study attempted to elucidate the relationship between PM deposition to leaves and ambient PM concentration reductions by estimating deposition velocity via the quantification of particle deposition to a hedge. Particle deposition velocity (V_d) depends on particle size and density, meteorological conditions (e.g. atmospheric stability and heights, wind speed, humidity, precipitation), vegetation canopy morphology, leaf surface characteristics and ambient concentrations of other pollutants (Beckett et al., 2000; Freer-smith and Taylor, 2000; Gallagher et al., 1997; Nowak et al., 2013; Zhang et al., 2001; Janhall, 2015 and articles herewith it). Findings from the present study underscore the plant species- and site-specific nature of PM deposition velocity to GI. We obtained deposition velocities (mean $\pm$ standard error) of $0.009 \pm 0.0008 \text{ cms}^{-1}$, $0.20 \pm 0.0134 \text{ cms}^{-1}$ and $0.49 \pm 0.0403 \text{ cms}^{-1}$ for PM size ranges PM_1 , $\text{PM}_{2.5-1}$, and $\text{PM}_{10-2.5}$, respectively. Figure 7 presents a comparison of acquired V_d values against those from wind tunnel experiments and recent model-predicted values, which displayed reasonable agreement. The difference between measured and modelled V_d may be due to model sensitivity in capturing complex physical and chemical processes involved in ambient PM capture by GI (Chen et al., 2017b). Moreover, this study's results aligned with those of previous studies, which reported V_d values for vegetation in the range of $0.02\text{-}10 \text{ cms}^{-1}$ and an average value of $\sim 1 \text{ cms}^{-1}$, with extreme values reaching up to $20\text{-}30 \text{ cms}^{-1}$ (Freer-Smith et al., 2005; Litschike and Kuttler, 2008; Zhang et al., 2001).

Further insights into PM deposition to leaves of the hedge can be gained by plotting the rate of PM deposition (Equation (3)) during the day and comparing this rate with the difference between INF and BHD monitoring points at the GI and clear area sites. As PM deposition is proportional to ambient concentration close to the GI (Popek et al., 2015; Weber et al., 2014), the PM deposition rate was predominantly higher in the morning traffic peak hours, followed

by the evening peak hours (SI Figure S5). These higher rates of a deposition provided only an indication of hourly variation in PM deposition because the study did not collect complete data during entire traffic peak hours (07:00-10:00h and 16:00-19:00h at this site). Higher concentrations and lower dispersion during morning peak hours may have resulted in higher PM deposition and thus led to a larger difference between INF and BHD monitoring points at GI compared to clear-area. As with the relative change (discussed in Sections 3.1), the absolute difference between INF and BHD monitoring points at the GI site was consistently higher than that of the clear area, positively indicating a combination of dispersion and deposition in the reduction of PM by GI (SI Figure S5).

3.4 Elemental analysis of deposited particles

Elemental analysis was carried out on a total of 2741, 2731, 2471, and 2674 particles deposited on leaves collected from sampling points $F_{1.5m}$, $F_{0.6m}$, $B_{1.5m}$, and $B_{0.6m}$, respectively. This study identified the following elements from the deposited particles: O, Na, Mg, Al, Si, P, S, Cl, K, Ca, Ti, Cr, Mn, Fe, Ni, Cu, Mo, Ba. These findings are in line with those of similar, previous studies (Mori et al., 2018; Ram et al., 2014; Sgrigna et al., 2015; Shao et al., 2018; Shi et al., 2017; Song et al., 2015). Particles were classified according to their elemental composition as ‘natural,’ ‘vehicle’ and ‘unclassified/agglomerates,’ as discussed in Abhijith and Kumar (2019) (Figure 8). ‘Natural’ particles primarily consist of soil and dust (Si, Ca, Al, Mg, Fe, K, S and P) (Jancsek-Turóczy et al., 2013; Ottel et al., 2010; Panda and Shiva Nagendra, 2018; Shao et al., 2018; Shi et al., 2017; Song et al., 2015), whereas particles of the ‘vehicle’ classification include elements that originate from the exhaust and non-exhaust sources, primarily comprising iron and rust, and traces of Ba, Cr, Mn, Cu, Ni and Ti (Gonzlez et al., 2017; Mazziotti Tagliani et al., 2017; Shi et al., 2017; Song et al., 2015; Weerakkody et al., 2018b). The agglomerated ‘natural’ and ‘vehicle’ particles are denoted as ‘unclassified/agglomerates.’ The percentage of particles of each classification is

provided in Figure 8, and summary statistics of percentage weight with regards to the elemental composition of particles is provided in SI Table S6.

At all four sampling points, most (>64%) of the deposited particles were found to belong to the ‘natural’ classification. This fraction was as high as 80% (+15% difference) for leaves sampled from the backside the of the hedge ($B_{1.5m}$ and $B_{0.6m}$) rather than the traffic-facing front side ($F_{1.5m}$ and $F_{0.6m}$). The comparison of sampling heights on both sides of the hedge showed a negligible difference in the proportion of ‘natural’ particles. Particles of the ‘vehicle’ and ‘agglomerates’ classifications were –8% to –12% and –5% to –8%, respectively, lower at the backside of the hedge than at the traffic-facing side. This may indicate fewer harmful particles in the ambient air behind the hedge, adding weight to the findings of Abhijith and Kumar (2019). They also reported higher quantities of ‘natural’ particles in the ambient air behind the hedge than in front, resulting in higher percentages of ‘natural’ particle deposition on the backside of the hedge. While comparing sampling heights, the percentages of the agglomerates and vehicle portions were closer in the backside of hedge alike traffic facing side. Most traffic-originated particles were deposited on the traffic-facing side, indicating and a positive barrier effect of GI in improving air quality.

4. Summary and conclusions

We compared ambient PM concentration and PM deposition to leaves, deriving deposition velocity for the hedge during the autumn period. We observed a greater PM reduction between monitoring points BHD and INF at the GI site than at the adjacent clear area, and the trend of PM reduction followed $\Delta PM_1 > \Delta PM_{10} > \Delta PM_{2.5}$. Counting of particles was identified as an effective method to quantify PM deposition on leaves compared to leaf washing technique. A higher deposition of all particle sizes was reported on leaves at the back of the hedge than at the traffic-facing front side, with the highest density reported for

PM of the fine (PM_{10}) size fraction. Moreover, PM density followed an order of $PM_{10} > PM_{2.5-10}$, comprising 66%, 29% and 5% of total deposited particles, respectively. In addition, adaxial leaf surfaces captured up to three times more particles than abaxial leaf surfaces. The mean deposited PM mass on leaves followed an opposite trend to that of PM density; i.e. $PM_{10} < PM_{2.5} < PM_{2.5-10}$. Interestingly, leaf sampling height significantly influenced PM deposition. Significant variation in PM density was observed on leaf samples from the traffic-facing front side of the hedge, whereas negligible variation in PM density was observed on the leaves from the backside of the hedge. The PM deposition rate was higher during morning traffic peak hours. Elemental analysis of deposited particles found most particles from natural sources (i.e. excluding particles classified as 'vehicle' and 'agglomerates') at all sampling points. A comparatively higher percentage of traffic-generated particles were captured by leaves on the traffic-facing side of the hedge than the backside of the hedge. This study provided insights into the relative contribution of deposition and dispersion in the reduction of PM concentrations by GI. Nevertheless, combined experimental and modelling investigations are required to depict real-time PM reductions arising from GI-induced dispersion and deposition. While our combined leaf deposition analysis and portable set-up for ambient PM measurement enabled a description of PM capture and reduction by a hedge, it is important to investigate and assess the influences of seasonal variation to develop comprehensive GI implementation guidelines.

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697 **List of Figure**

698

699 **Figure 1.** Aerial view of the study location, showing the clear area (1) and hedgerow (2).

700 Central section shows the sites at street level, with measurement points marked as stars.

701 Section on the right includes schematic diagrams of hedge and road dimensions and height of

702 monitoring points.

Figure 2. Boxplots of PM concentration behind (red) and in front (green) measurement points at GI site and clear area (CLR) for (a) PM_{10} , (b) $\text{PM}_{2.5}$, (c) $\text{PM}_{10-2.5}$ (coarse fraction), and (d) PM_1 . The blue dot shows the mean concentrations.

707

708 **Figure 3.** Percentage differences in PM concentration at the GI and clear area sites. The
 709 shaded bar and solid bars represent the clear area and GI sites, respectively.

Figure 4. Box plots and hourly average relative concentration of PM (PM_{BHD}/PM_{INF}) at GI (green: box, dots and line) and Clear area (red: box, dots and line). The dashed line shows $PM_{BHD}/PM_{INF} = 1$.

714

715 **Figure 5.** Bar plots showing size-segregated PM densities on leaves from the front side of hedge (at
 716 1.5m height, F_{1.5}; at 0.6m height, F_{0.6}) and backside of the hedge (at 1.5m height, B_{1.5}; at 0.6m height:
 717 B_{0.6}). The top row shows abaxial and adaxial PM densities of coarse (a) and fine (b, c, d) PM
 718 fractions, whereas bottom row displays overall PM densities of respective size ranges (e, f, g, h).

719 **Figure 6.** Bar plots showing size-segregated PM deposition on leaves from the front side of hedge
 720 (at 1.5m height, F_{1.5}; at 0.6m height, F_{0.6}) and backside of the hedge (at 1.5m height, B_{1.5}; at 0.6m
 721 height, B_{0.6}) for (a) PM₁₀, (b) PM_{10-2.5}, (c) PM_{2.5}, (d) PM_{2.5-1}, and (e) PM₁.
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Figure 7. Comparison of obtained deposition velocity with those from previous experimental values (Zhang et al., 2014) and modelling study (Giardina and Buffa, 2018).

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Figure 8. Percentage of samples identified of each elemental composition group from total particles deposited on leaves at: (a) front side of hedge at 1.5m height / $F_{1.5}$; (b) front side of hedge at 0.6m height / $F_{0.6}$; (c) backside of hedge at 1.5m height / $B_{1.5}$; and (d) backside of hedge at 0.6m height / $B_{0.6}$.

List of Tables

Table 1. Percentage difference in PM density and deposition between sampling heights and sides of the hedge. In the height column, PM deposition and density on leaves from 1.5 m are compared with leaves from 0.6 m taking 1.5 m height as a reference. In the hedge sides column, PM deposition and density on leaves from same heights on either side are compared taking a front side as a reference.

	Size fraction	Height 1.5 m vs 0.6m		Hedge Sides Front vs Back	
		Front	Behind	1.5m vs 1.5m	0.6m vs 0.6m
% PM Deposition	PM ₁₀	-5	5	-22	-14
	PM _{10-2.5}	4	4	-29	-29
	PM _{2.5}	-33	7	-27	11
	PM _{2.5-1}	-36	7	-29	11
	PM ₁	-8	9	-12	6
% PM density	PM _{10-2.5}	-16	9	-34	-5
	PM _{2.5}	-50	8	-56	4
	PM _{2.5-1}	-37	8	-34	10
	PM ₁	-58	8	-66	3

Scale

- Very Low (<10%)
- Low ($\geq 10\%$ < 20%)
- Moderate ($> 20\%$ $\leq 30\%$)
- High ($> 30\%$ $\leq 50\%$)
- Very High ($> 50\%$)

Research highlights

- ▶ Presence of GI leads to PM reduction behind them to as: $\Delta PM_1 > \Delta PM_{10} > \Delta PM_{2.5}$.
- ▶ Magnitude of percentage PM reduction behind GI is lower in autumn than spring- summer
- ▶ PM_1 dominates in particles deposited on leaves of the hedge
- ▶ PM deposition to leaf varies according to sides and heights of the hedge
- ▶ Majority of traffic-generated particles are captured on leaves on the traffic facing side

Declaration of interests

☒ 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.

☐ The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: