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JES JOURNAL OF ENVIRONMENTAL SCIENCES

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Chemical characterisation of particulate matter in urban transport modes

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ARTICLE INFO

Article history:
Received 11 May 2020
Revised 3 July 2020
Accepted 6 July 2020
Available online 23 July 2020

Keywords:
Traffic
Commuting
Air pollution
Particle chemistry
Indoor-outdoor interplay
Inhaled dose

ABSTRACT

Traffic is a main source of air pollutants in urban areas and consequently daily peak exposures tend to occur during commuting. Personal exposure to particulate matter (PM) was monitored while cycling and travelling by bus, car and metro along an assigned route in Lisbon (Portugal), focusing on PM2.5 and PM10 (PM with aerodynamic diameter <2.5 and 10 μm, respectively) mass concentrations and their chemical composition. In vehicles, the indoor-outdoor interplay was also evaluated. The PM2.5 mean concentrations were 28 ± 5, 31 ± 9, 34 ± 9 and 38 ± 21 μg/m^3 for bus, bicycle, car and metro modes, respectively. Black carbon concentrations when travelling by car were 1.4 to 2.0 times higher than in the other transport modes due to the closer proximity to exhaust emissions. There are marked differences in PM chemical composition depending on transport mode. In particular, Fe was the most abundant component of metro PM, derived from abrasion of rail-wheel-brake interfaces. Enhanced concentrations of Zn and Cu in cars and buses were related with brake and tyre wear particles, which can penetrate into the vehicles. In the motorised transport modes, Fe, Zn, Cu, Ni and K were correlated, evidencing their common traffic-related source. On average, the highest inhaled dose of PM2.5 was observed while cycling (55 μg), and the lowest in car travels (17 μg). Cyclists inhaled higher doses of PM2.5 due to both higher inhalation rates and longer journey times, with a clear enrichment in mineral elements. The presented results evidence the importance of considering the transport mode in exposure assessment studies.

Introduction

Several epidemiological studies have found associations of human exposure to traffic-related air pollution with adverse cardiovascular and respiratory outcomes (Escamilla-Nuñez et al., 2008; Kubesch et al., 2015; Peters et al., 2004; Samoli et al., 2016; Vahedian et al., 2017; Wu et al., 2010) or mortality (Beelen et al., 2008; Madsen et al., 2012). Although the majority of the studies commonly relate health effects with the air pollutants concentrations from fixed air quality monitoring stations (Özkaynak et al., 2013 and references therein), the real personal exposure varies greatly with location, activity and time spent on each activity (Bekö et al., 2015;

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https://doi.org/10.1016/j.jes.2020.07.008
1001-0742/

Buonanno et al., 2013a; Cunha-Lopes et al., 2019; Steinle et al., 2015). The use of data from fixed site monitoring stations generally underestimates commuter exposures, principally on or close to heavily trafficked roads (Knibbs et al., 2011 and references therein). Traditionally, air pollution monitoring uses expensive, complex and stationary equipment, with electrical power dependence, which do not suit the need for personal exposure assessment. This technical difficulty is gradually being overcome by the fast technological development of portable air pollution monitoring equipment, which contribute by providing information relative to exposure assessment and subsequent risk mitigation (Engel-Cox et al., 2013; Morawska et al., 2018; Steinle et al., 2015, 2013).

Typically, people in Europe spend around 5–8% of their daily time commuting (including walking) (Faria et al., 2020; Hänninen et al., 2005), yet even this short amount of time may contribute substantially to the total exposure to particulate matter (PM) (Künzli et al., 2000). High PM concentration levels have been indicated in different transport modes (car, bus, metro, motorbike, bicycle and pedestrian) (Cepeda et al., 2017; de Nazelle et al., 2012; Fernández-Iriarte et al., 2020; Martins et al., 2016a; Moreno et al., 2015a; Okokon et al., 2017; Rivas et al., 2017). Traffic-related particles can enter into the vehicles and expose both driver and passengers to high PM concentrations. Commuting accounts for a short fraction of the daily period, nevertheless, the contribution to the daily inhaled airborne pollutants may be disproportionally high, depending on local pollution and traveling mode. Active transport (walking and cycling) brings undoubted health benefits to commuters, by increasing their activity level. However, this high physical activity increases the breathing rate and hence, the intake of air pollutants. The peak levels of exposure to air pollutants during a commuting period of 1-hr or less, may pose relevant health risks (Michaels and Kleinman, 2000). Therefore, exposure assessment to traffic-related pollution during commuting remains a major public health issue in urban areas with heavy traffic owing to their proximity to emission sources, deserving increased attention.

Road traffic is considered the main source of primary PM (both exhaust and non-exhaust particles) and precursor gases (e.g., NOx, SO_2 and volatile organic compounds) in the urban areas, and especially at roadside locations (Amato et al., 2015; Charron et al., 2007; Gaba and Iordache, 2011; Querol et al., 2004). Non-exhaust particles are typically associated with abrasive emissions from brake, road, and tyre wear, and road dust resuspension (Thorpe and Harrison, 2008). In recent years, the introduction of increasingly stringent European emission standards has achieved significant reductions in exhaust emissions from road traffic, while the non-exhaust emissions remain uncontrolled. Indeed, increased attention is focusing on non-exhaust emissions as they are already considered the main PM source in several urban areas (Amato et al., 2014; Bukowiecki et al., 2010; Denier van der Gon et al., 2013; Font and Fuller, 2016). Vehicle non-exhaust sources are mainly associated with the emission of PM with aerodynamic diameter less than 10 μm (PM10) (Harrison et al., 2012; Thorpe and Harrison, 2008), while vehicle exhaust emissions produce finer particles, such as PM with aerodynamic diameter less than 1 μm (PM1) and nanoparticles (Kumar et al., 2010; Kumar and Goel, 2016). Non-exhaust emissions such as brake and tyre wear are a major source of airborne trace metals in the urban environment, surpassing industrial sources in areas of traffic influence. The abrasion of road surfaces, commonly made of either concrete or asphalt, is likely to release PM of mineral origin (Tervahattu et al., 2006). Resuspended road dust consists of material from traffic related sources, such as vehicle abrasion debris and exhaust pollutants, and non-traffic related material (Pakbin et al., 2011; Thorpe and Harrison, 2008).

Assessing human exposure to PM in urban transport environments is challenging, as PM concentrations and particle sizes vary depending on the route, traffic intensity, commuting mode, fuel type, in-vehicle ventilation mode, distance from the source and time of day (e.g. Asmi et al., 2009; Cepeda et al., 2017; Correia et al., 2020; Fernández-Iriarte et al., 2020; Moreno et al., 2015a; Zhu et al., 2002; Zuurbier et al., 2010). These variables are very dynamic among the regions of the globe, as they are influenced by meteorology, driving behaviour and existing infrastructure.

Most of the studies determining the particle exposure in transport modes have focused on the real-time PM mass and number concentrations and respective health effects, neglecting its chemical composition (e.g. Briggs et al., 2008; de Nazelle et al., 2012; Johansson et al., 2017; Kaur et al., 2007; Knibbs et al., 2011; Okokon et al., 2017); this is largely due to the lack of a portable, robust device to collect particles. The present study characterises personal exposure to PM chemical components in various transport modes (bicycle, bus, car and metro) along an assigned route in Lisbon, Portugal. The specific aims of this work were (1) to assess PM concentration differences among the four modes of transportation, (2) to identify the chemical components of PM, (3) to compare the indoor-to-outdoor concentrations of PM and its chemical constituents in transport vehicles, and (4) to evaluate the contribution of commuting to total daily dose. To the best of the authors' knowledge, this is the first study evaluating the PM's chemical components concurrently in the outside of the vehicle in order to obtain the indoor-to-outdoor relationship. This approach provides useful data for exposure assessment models.

1. Method

1.1. *Study area*

Lisbon is the largest city of Portugal and the continental Europe's westernmost capital city. It has a population of about 2.8 million inhabitants, spread over about 3015 km^2. Lisbon centre is located at the point where the Tagus River flows into the Atlantic Ocean coast and it is surrounded by seven hills. The enclosed location inhibits ventilation which, when combined with predominantly narrow streets and scarce green areas, causes the accumulation of pollutants. Traffic emissions are considered one of the main contributors to the large PM concentrations observed in the city (Almeida et al., 2009a, 2009b). According to the annual report "Traffic Index 2018", Lisbon ranked as the most congested city in the Iberian Peninsula (based on TomTom navigation data). In 2017, the transport fleet in the city included 366,671 vehicles; of which 80% were passenger cars, 6% motorcycles, 0.4% buses, and 14% other types (ASF, 2018). Moreover, the public transport system is also composed of an electrical underground metro system. Major air and maritime transport infrastructures may also contribute to the air pollution within the city, i.e. the largest airport in Portugal and the port of call for cruises, respectively. The city is also significantly affected by marine aerosol (Almeida et al., 2013) and North African mineral dust (Almeida et al., 2008) due to its geographical position. Air pollution events in the city are frequently observed, especially in winter, due to adverse meteorological conditions, such as limited dispersion and atmospheric thermal inversions (Alves et al., 2010).

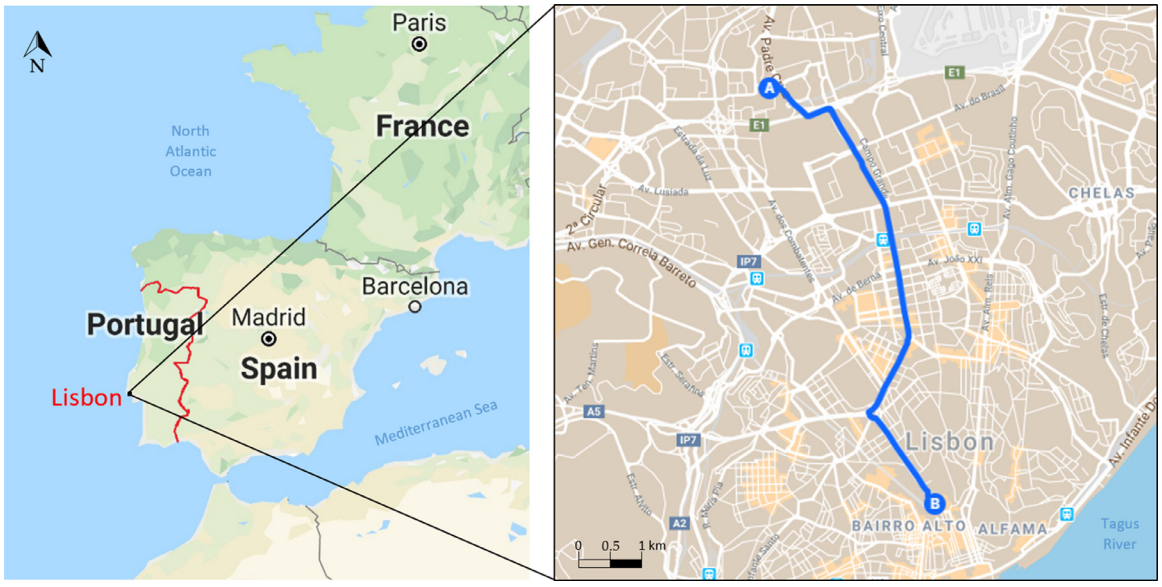

Fig. 1 – Selected route - starts in Telheiras (A) and ends in Praça dos Restauradores (B).

1.2. *Sampling design*

The definition of a fixed route is an effective and simple approach to compare the personal exposure to air pollutants in different transport modes (Suárez et al., 2014). Thus, the field measurements were carried out in the four most common modes of transport used in Lisbon (bicycle, bus, car and metro) along an assigned route, covering a distance of 6.7 km by road. The route starts in Telheiras, a residential area, and ends in Praça dos Restauradores located in the downtown area (Fig. 1). This route was selected because it represents a path of many typical commutes in Lisbon city centre and comprises infrastructure for all four transport modes, including bike paths, bus lanes and underground metro. It is characterised by a high variety of conditions, covering different traffic densities, street configurations (e.g. bike path within the road or in the sidewalk), dispersion conditions, and vehicle fleet composition (existence of low emission zones).

Measurements were performed over a total of 21 weekdays between June and October 2018 at 5 different daytime periods (starting at 8 hr, 10 hr, 13 hr, 18 hr and 20 hr). The measurements period was of 3 days each for the bicycle and metro modes, whereas for bus and car modes the measurements were conducted during 5 and 10 days, respectively. More trips in car were monitored since it is the most prevalent transport mode in Lisbon. The measurements were carried out in cars of different brands and fuel type, and using diverse ventilation modes to broaden the representativeness of the sample regarding exposure conditions, as it happens under real conditions. No buses that ran the entire length of the study route exist, so two bus lines were selected, and no measurements were carried out during the connection time at the bus stop. The ventilation conditions were not controlled in the public transport modes. The metro route involved travelling in three trains: green line from Telheiras to Campo Grande, yellow line from Campo Grande to Marquês de Pombal and blue line from Marquês de Pombal to Restauradores. The time spent waiting on the platforms and walking in the connecting corridors to make the changes were considered.

All study personnel were non-smokers. A manual record with the registration of the movements time and indication of the locations affecting the exposure during commuting was performed. The average duration of a return trip was 44, 57, 58 and 77 minutes for car, metro, bus and bicycle mode, respectively. The measurements were performed under no air pollution events, such as low dispersion conditions, thermal inversions and African dust episodes.

In cars and buses, the measurements were performed in the indoor and outdoor air simultaneously. This approach allowed to evaluate the indoor-to-outdoor relationship between the air pollutants. The devices measuring outdoors were placed inside the vehicles with the air inlets outside of the window. The inlets were fitted with a protection plastic cup in a position opposite to the movement. An isolator was placed in the open window space to prevent the air exchange.

1.3. *Measurements and instrumentation*

1.3.1. *PM collection*

PM2.5 and PM10 samples were collected on 37 mm polytetrafluoroethylene filters (PTFE) using two Personal Environmental Monitors (PEM; no. 761-203B for PM2.5 and no. 761-200B for PM10, SKC Inc., USA), which are compact sampling devices with a single stage impactor. The PEM was connected to an air suction pump (Leland Legacy, SKC Inc., USA), operating at a flow rate of 10 L/min. The flow was checked at the beginning of each sample using a flowmeter (Bios Defender 510, MesaLabs, USA) and always set to within ±0.5% of the target flow rate. Each day prior to the sampling, the pump's internal clock was reset and the PEMs were cleaned and its impaction plate was greased. The measurements in each transport mode were repeated 5 times a day in order to obtain enough PM mass for posterior chemical analysis. The PM2.5 and PM10 samples represent the daily average mass concentrations for each mode of transport. No PM10 data were collected in the buses due to technical issues.

1.3.2. *Real-time BC concentrations*

Black carbon (BC) concentrations were measured with a portable micro-aethalometer (microAeth® Model AE51, AethLabs, USA), which has been commonly used in personal exposure assessment (Buonanno et al., 2013b; Cunha-Lopes et al., 2019; Moreno et al., 2015a; Rivas et al., 2016). This monitor

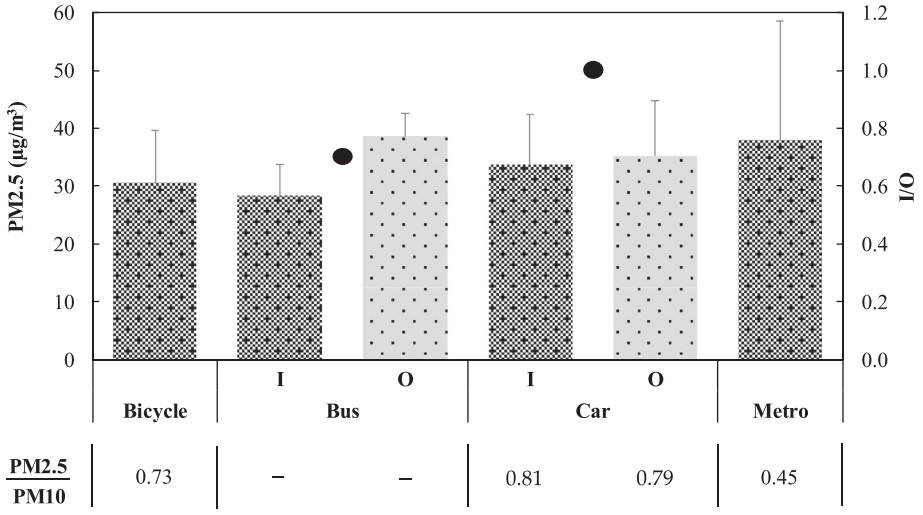

Fig. 2 – PM2.5 mean concentrations, standard deviations and respective PM2.5/PM10 ratios in the four commuting modes. Black dots represent the I/O ratios for PM2.5. Unavailable data for PM10 in the buses. I: Indoor; O: Outdoor.

determines BC concentration in real-time based on the Beer–Lambert law, by measuring the changes of absorption of light at the 880 nm wavelength by the aerosol sample continuously collected on a Teflon-coated borosilicate glass fiber filter. The effect of filter loading was minimised by replacing the filter strips whenever the equipment indicated as necessary. The monitor was operated at a flow rate of 100 mL/min and the time set to 10 sec, as recommended by the manufacturer for optimum performance in traffic and transportation measurement scenarios. The BC reported data are the daily average mass concentrations for each mode of transport. The performance of this portable monitor has been evaluated and showed a good agreement comparing with widely used stationary instruments (Cheng and Lin, 2013; Viana et al., 2015).

Under low BC concentrations or inadequate high time resolution of the measurements, the light absorption of the collected sample between timestamps may be lower than the changes in signal caused by instrumental optical and electronic noise, resulting in low signal to noise ratio. Ultimately, this can result in negative BC concentrations reported by the AE51 device (Cheng and Lin, 2013), and can be effectively improved by adopting the Optimised Noise-reduction Averaging (ONA) algorithm (Hagler et al., 2011). Therefore, the original data obtained in this study was post-processed with the ONA algorithm.

The instruments used were transported in a bag with the air uptake inlets placed at the commuter's breathing zone. The bag was carried on the back while biking or standing in the bus or metro. If the study personnel were seated, the bag was placed on the lap with inlets also at breathing zone. During car measurements, the instruments measuring inside were placed on the front passenger seat.

1.4. *Sample analysis*

PM mass concentrations were determined gravimetrically using a microbalance (R160P, Sartorius, Germany). The filters were weighed before and after sampling, after being equilibrated for at least 24 hr in a conditioned room (20°C and 50% relative humidity). After weighing, the sampled filters were stored in a freezer (≈ −15°C) until chemical analysis.

The X-Ray Fluorescence (XRF) technique was applied to determine concentrations of the following major and trace elements: Na, Mg, Al, Si, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, As, Sb, Ba and Pb. An Energy Dispersive X-ray spectrometer (ED-XRF) Laboratory Instrument (Epsilon 5, PANalytical, The Netherlands) was used (Manousakas et al., 2018). A detailed description of the analytical procedures is given by Popovicheva et al. (2019). The final ambient concentrations were obtained after the subtraction of the blank values from the corresponding sample concentrations.

1.5. *Statistical analysis*

The analytics software "STATISTICA" was applied to analyse the obtained results. The comparison between two paired groups was analysed by Wilcoxon Matched Pairs Test (differences between the pollutant concentrations in-vehicle versus outdoor), while Mann-Whitney U Test was applied to compare two independent groups (differences of the pollutant concentrations between transport modes). $p < 0.05$ indicated the statistical significance.

1.6. *Dose calculation*

To calculate the inhaled dose of PM2.5 and PM10 and of their respective chemical components per commuting mode, the inhalation rates used were based on the study developed by Zuurbier et al. (2009). Biking had the highest inhalation rates (23.5 L/min) due to the elevated physical activity associated to this commuting mode. The same inhalation rate (12.7 L/min) was considered for people commuting by bus and metro transport modes. Car transportation had an inhalation rate of 11.8 L/min. The inhaled dose (μg or ng of pollutant per daily commute) was calculated by the product between the mean pollutant concentration (μg/m^3 or ng/m^3) for each commuting type, the inhalation rate (m^3/hr) and the average of the time spent daily commuting (hr).

2. Results and discussion

2.1. *Variation of PM concentration by commuting mode*

Fig. 2 shows the PM2.5 mean concentrations and respective PM2.5/PM10 ratios in the four commuting modes. The lowest PM2.5 concentrations were found when travelling by bus

(28 μg/m^3: range 21–35 μg/m^3), with concentrations increasing for the bicycle journeys (31 μg/m^3: range 21–38 μg/m^3), the car (34 μg/m^3: range 24–52 μg/m^3), and the metro (38 μg/m^3: range 23–62 μg/m^3). The arithmetic mean PM concentrations and respective standard deviations are given in **Table S1** (supplementary data). In buses the lower PM2.5 mean concentrations obtained in comparison with the cars may be associated with the existence of recirculating air-conditioning in buses, as will be discussed further. Conversely, Moreno et al. (2015a) found the mean PM2.5 bus concentration to be 45 μg/m^3 — higher than in the other modes of transport studied (walking, tram and metro). The variations in PM2.5 concentrations among samples collected inside the cars may be associated with the use of diverse ventilation modes to increase the data representativeness, as described in section 2.2. Correia et al. (2020) verified that PM concentrations were greatly dependent on the ventilation mode used. They concluded that the car air filters coupled to the air ventilation system prevent partially the infiltration of particles from outdoor, leading to lower PM2.5 and PM10 concentrations. The highest indoor-to-outdoor ratios were observed in journeys with ventilation system turned off (Correia et al., 2020). Similarly, other studies have suggested that the in-vehicle filtration system might help to prevent the penetration of air pollution from the outdoor environment (Briggs et al., 2008; Muala et al., 2014; Yu et al., 2017).

The PM2.5 mean concentrations in the outdoor of both buses (39 μg/m^3: range 31–42 μg/m^3) and cars (35 μg/m^3: range 19–45 μg/m^3) were in the same range of values found when biking (31 μg/m^3: range 21–38 μg/m^3), as expected. The concentrations were slightly lower when biking probably explained in part by the fact that, in some fractions of the route, the bike path is separated from the roadway and thus less affected by the traffic-related pollution sources. For example, Kumar and Goel (2016) found that the exposure to on-road PM10 in-car when windows were fully open was up to ≈7-fold higher than that for pedestrians at the traffic intersections, which evidences that exposure varied with distance from motorised traffic.

PM2.5 mean concentrations in-vehicles (cars and buses) were lower than those in the respective ambient air (**Fig. 2**). On average, the PM2.5 concentrations inside the bus were 26% lower than outside, while for car, the difference was only of 4%. This higher difference in the indoor-to-outdoor PM2.5 concentrations in the buses was probably due to the existence of recirculating air-conditioning, that limits air exchange rates and promotes particle losses to surfaces, as mentioned by de Nazelle et al. (2012).

When biking and travelling by car, the PM2.5/PM10 ratio varied between 0.73 and 0.81 (**Fig. 2**), evidencing that the particles in urban traffic environment are mostly fine. These fine particles are mainly originated from exhaust sources, while the coarse particles are generally associated with non-exhaust sources such as road surface abrasion and wear of brakes and tyres. However, several studies have revealed that wear particles can also contribute considerably to the fine and ultrafine fractions (Keuken et al., 2010; Kukutschová et al., 2011; Kwak et al., 2013; Sanders et al., 2003; von Uexküll et al., 2005).

The greatest variability between PM2.5 concentrations was observed when travelling by metro, which was also observed in the commuting study conducted in Barcelona (Moreno et al., 2015a). The PM2.5/PM10 ratio in the metro was 0.45, which is consistent with a study carried out on a platform of Prague underground metro (ratio = 0.44) (Cusack et al., 2015). Cusack et al. (2015) performed measurements when the metro was both inoperative and in operation, and verified that although PM levels were high during both periods, the coarse fraction was more dominant during operating hours. Previous studies have indicated that the ultrafine particles present on metro stations are mostly introduced from the outdoor urban environment (Mendes et al., 2018; Reche et al., 2017), while the evolution of particles with aerodynamic diameter (Dp) between 0.3 and 10 μm was associated with activities of the metro service (Reche et al., 2017). According to Moreno et al. (2015b), the coarser fraction of PM, with Dp between 5 and 10 μm, is usually related to mineral particles. An important metro source of mineral particles is the weathering and erosion of ballast and construction materials, occasionally enhanced by construction or maintenance works in metro systems (Martins et al., 2017; Minguillón et al., 2018). Additionally, mineral particles can be carried in from outdoor by the commuters and through air exchange (Martins et al., 2017). The chemical characterisation of the particles collected not only during commuting by metro but also in the other transport modes will be discussed further below.

2.1.1. *Black carbon*

Black carbon (BC) has been considered a better tracer of traffic-related emissions than PM mass concentration (Reche et al., 2011). The BC mean concentrations are displayed in **Fig. 3**. The highest BC mean concentrations were obtained when travelling by car (6.1 μg/m^3: range 2.6–8.4 μg/m^3), followed by metro (4.5 μg/m^3: range 3.8–5.1 μg/m^3), cycling (4.2 μg/m^3: range 3.5–5.7 μg/m^3), and the lowest was registered inside the bus (3.0 μg/m^3: range 1.1–5.9 μg/m^3). The arithmetic mean BC concentrations and respective standard deviations are given in **Table S1** (supplementary data). The bus was the cleanest of the four transport modes with respect to BC, as observed for PM. In the study conducted in Barcelona by de Nazelle et al. (2012), they also reported that the highest BC concentrations were obtained when travelling by car compared to other transport modes (walking, biking and bus). This probably results of the closest proximity to the vehicle's exhaust system, as demonstrated by de Nazelle et al. (2012) and Targino et al. (2016). High BC levels in the urban environment are an indicator of road traffic pollution, specially from diesel vehicles. Inside the cars, the BC concentrations showed to be dependent on the type of ventilation and type of cabin air filter, as discussed in Correia et al. (2020). They observed lower BC concentrations when ventilation was turned off and that activated carbon filters are more efficient retaining BC than traditional filters.

The BC concentrations in the outdoor of both buses (4.7 μg/m^3: range 2.5–6.8 μg/m^3) and cars (7.7 μg/m^3: range 3.5–12.2 μg/m^3) were slightly higher than those found when biking (4.2 μg/m^3: range 3.5–5.7 μg/m^3). Recently, Amato et al. (2019) demonstrated that the BC concentration decreases exponentially with increasing distance from road edge. Thus, as in some fractions of the route the bike path is separated from the roadway, it is less affected by the traffic-related pollution sources.

BC mean concentrations in-vehicles (cars and buses) were lower than those in the ambient atmosphere (**Fig. 3**), as observed for PM. On average, the BC concentrations inside the bus were 36% lower than outside, while for car the difference was 21%. These indoor-to-outdoor differences were more relevant for BC than for PM2.5, which may be attributed to the existence of PM2.5 indoor sources, such as resuspension of settled particles and possible particle generation. Moreover, the BC concentrations in the outdoor of cars showed to be more variable than in buses (**Fig. 3**). This variability may be related to changes in traffic emissions and weather conditions, as suggested by Targino et al. (2016). The traffic emissions are dependent on several variables, such as traffic stream properties (density and flow), road typology and vehicle fleet characteristics (exhaust emission standards). The density and flow of the

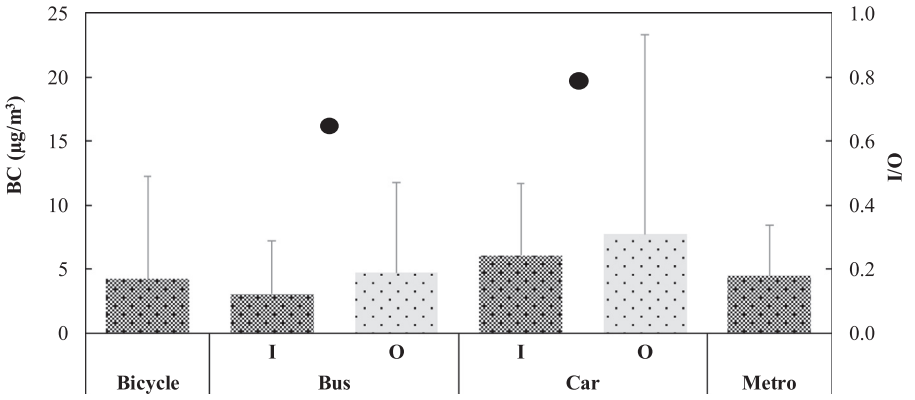

Fig. 3 – BC mean concentrations and standard deviations for the four commuting modes. Black dots represent the I/O ratios. I: Indoor; O: Outdoor.

traffic may particularly affect more the pollutant concentrations in cars than in buses. Although both transport modes circulate in close proximity to the exhaust of other vehicles, the buses run on dedicated lanes being less prone to traffic jam impacts. Additionally, traffic congestions have been shown to rise driver's exposure to airborne pollutants up to 29 times, compared to fluid traffic flow (Goel and Kumar, 2015).

Considering the results for both car and bus journeys, the BC concentrations were not correlated with the values of PM2.5 (**Fig. S1**, supplementary data). The main sources of PM2.5 in traffic-related environments are non-exhaust particles associated not only with the abrasion of brakes, tyres and road surface, and subsequent road dust resuspension (Amato et al., 2014; Bukowiecki et al., 2010; Denier van der Gon et al., 2013; Font and Fuller, 2016), but also with formation and growth of ultrafine particles from secondary sources (Ahlm et al., 2012). Incomplete combustion of fossil fuels is the major environmental source of BC pollution in urban areas. Thus, an enhancement of PM2.5 will not necessarily be reflected in the BC concentrations, as also observed by Targino et al. (2016).

In terms of BC, travelling by car is a less attractive option than taking public transport modes. The metro commutes are the less attractive when considering PM concentrations, probably due in part to the high ferruginous nature of metro particles as explained by Moreno et al. (2015a). Numerous studies have indicated Fe as the most abundant chemical element constituting metro PM (e.g. Figueroa-Lara et al., 2019; Martins et al., 2016a; Van Ryswyk et al., 2017). This high amount of ferruginous particles lead to artificially high BC concentrations measured by the micro-aethalometer monitor (Moreno et al., 2015a), therefore BC levels are overvalued. Moreover, it is important to note that the metro trains are powered by electricity and, thus, it is somewhat unexpected to find high levels of BC in the metro system since there are no known combustion sources producing BC. However, studies have revealed as possible sources of BC in metro systems the diesel-powered trains used for maintenance works and most importantly the abrasion of C-bearing brakes and power supply materials (Martins et al., 2016a; Moreno et al., 2015b). Carbonaceous aerosol from outdoor may also enter the metro area through ventilation (Martins et al., 2017).

2.1.2. *Major and trace elements*

The mean concentrations of the chemical elements analysed in the PM2.5 and PM10 samples are displayed in **Fig. 4**. The chemical characterisation of the air breathed when commuting is an uncommon content in this type of studies. Most of the existing commuter exposure studies have focused on real-time PM measurements and not on its chemical constituents, which can be associated to the lack of portable devices to collect particles. Moreover, for chemical analysis it is necessary to repeat each commuting journey several times to collect enough particle mass, as also indicated by Moreno et al. (2015a) and Fernández-Iriarte et al. (2020). To the best of the authors' knowledge, this is the first study covering the topic of personal exposure to chemical components using different commuting modes in Lisbon. The results of this study demonstrate interesting differences in personal exposure to PM chemical components depending on the transport mode used. The arithmetic mean concentrations of the elemental components and respective standard deviations are given in **Table S1** (supplementary data).

Remarkable differences can be identified between the bicycle, bus and metro samples, in terms of their content in S, Fe, Cu and Zn, as will be discussed below. The car mode samples were not comparatively enriched in any chemical element (**Fig. 4**).

Considerable high amount of mineral related elements (sum of Fe, Si, Ca, Ti, K, Al and Ba) were observed when commuting by metro (11.9 μg/m^3 in PM2.5), bus (3.7 μg/m^3) and bicycle (4.0 μg/m^3). These results may be associated with the ambient air influence and, in the case of the buses, also affected by the resuspension of particles, created by the passengers' movement and the air flowing in and out when the doors are open. The mean concentration of mineral elements in the PM2.5 samples collected in the outdoor of the buses (4.4 μg/m^3) and cars (3.6 μg/m^3) were in the same range of values observed when biking. The mineral concentrations in car cabin were on average one third of those found in the outdoor environment for PM2.5.

In the metro microenvironment the air has a chemical composition very different, with a dominant content of ferruginous particles. On average, the relative contribution of Fe to the bulk PM2.5 in the metro was 28% (40% if Fe_2O_3 is considered). The Fe element was 9 times more abundant in the metro microenvironment than in the bicycle, or 26 times more than in the car. This high Fe abundance was accompanied by the presence, to a lesser extent, of trace elements such as Cu, Ba, Cr, Si, Mn, Zn and Pb. Such elements are associated with the abrasion of rails, wheels, brake pads, catenaries and pantographs (Font et al., 2019; Martins et al., 2016b; Minguillón et al., 2018). Mineral particles found in the metro microenvironment may result from the resuspension of particles generated by weathering of construction material, and

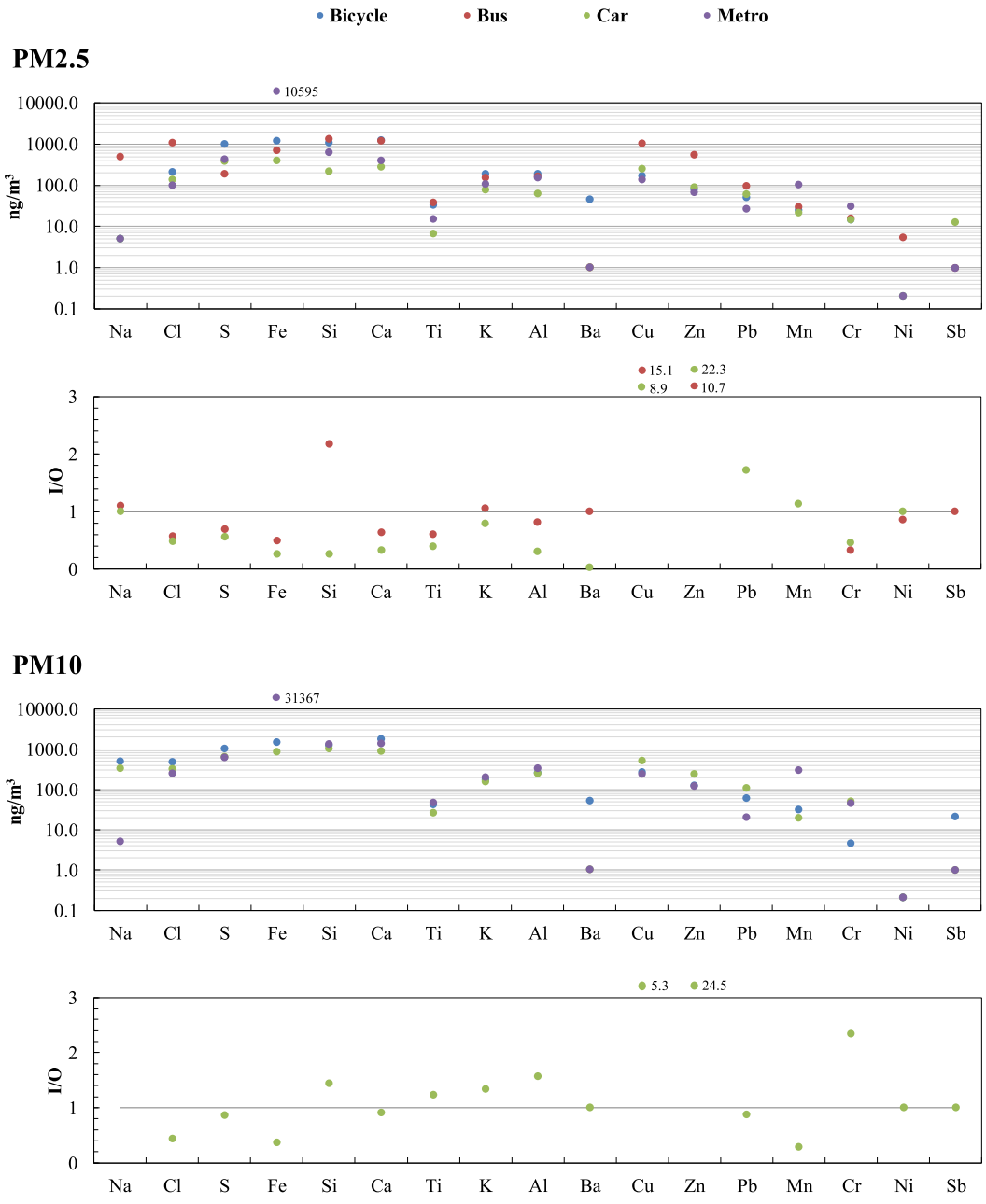

Fig. 4 – Mean concentrations of chemical components in the four commuting modes for both PM2.5 and PM10, and respective I/O ratios. Unavailable data for PM10 in the buses.

debris of occasional maintenance works. Moreover, some of these particles can also be carried in from outdoor by the passengers and through air exchange (Martins et al., 2016a).

In buses, the inhalable aerosol may also contain its own distinctive trace element mixture, with a clear enrichment in Cu and Zn, besides the relatively high concentrations of mineral related elements such as Si, Fe, Ca, Al, K and Ti (Fig. 4). Previous studies have shown that the dust resuspended from roadways is enhanced with elements emitted from several sources such as tailpipe emissions, brake wear and tyre wear (Amato et al., 2014 and references therein). The analysis of the exhaust of diesel engines indicated the presence of considerable content of Al, Ca, Fe, Si, Mg, Cu and Zn (Loyola et al., 2009; Wang et al., 2003; Weber et al., 2000), evidencing the relation between these metals and diesel combustion. The addition of metal-based additives containing Zn and Mg, anti-wear agents (Zn based) and detergents (Ca and Mg based) to fuels and lubricating oils has also been reported (Lim et al., 2007; Lyyränen et al., 2002; Wang et al., 2003; Weckwerth, 2001), influencing the size of particles emitted and their chemical composition (Lim et al., 2007). Moreover, the brake and tyre wear emissions contain significant amounts of metal, including Fe, Zn and Cu (Councell et al., 2004; Ochoa Gonzalez et al., 2016; Pant and Harrison, 2013). Other metals such as Ba, Mn, Ni, Sb, Sn, Cd, Cr, K, Ti and Pb have also been considered as key tracers for brake and tyre wear (Kukutschová et al., 2011; Pant and Harrison, 2013). The chemical composition of brake linings

and tyres is very dependent on the manufacturer and brand (Denier van der Gon et al., 2013; Pant and Harrison, 2013).

In-vehicle (merged data of cars and buses) the Fe, Zn, Cu, Ni and K were highly correlated between each other ($0.82 < R < 0.99$; **Table S2**, supplementary data) suggesting their common brake and tyre wear origin. This finding confirms that pollutants can enter the vehicle cabin not only through windows and doors but also through air vents and other openings in the vehicle enclosure. Passenger vehicles are usually equipped with disc brakes at the front and either disc or drum brakes at the rear. Since front brakes withstand most of the breaking effort (70%) (Hutchings and Shipway, 2017), the wearing of breaking materials is also expected to be higher at the front. The particles released from the abrasion of the front brakes and tyres are more likely to enter in the vehicle cabin, resulting in elevated I/O ratios of Cu and Zn for buses and cars (Fig. 4). Thus, these elements when enter in-cabin tend to accumulate, while, in the outdoor atmosphere, they are more easily dispersed. In the outdoor PM2.5 samples collected concurrently, the correlation coefficients between these metal elements (Fe, Zn, Cu, Ni and K) ranged from 0.17 to 0.81 (**Table S3**, supplementary data). As expected, the lowest correlations were observed in the outdoor because there are relevant extra particle sources, such as other traffic-related sources, such as tailpipe emissions, road wear and dust resuspension, and non-related traffic emissions (e.g. commercial and industrial processes).

On average, inside the buses the concentrations of Fe, Zn, Cu, Ni and K were higher than in-cars (Fig. 4). Buses usually stops more frequently than cars, which enhance the emission of brake and tyre wear particles.

The wear debris from brakes and tyres released into the air depends essentially on a) bulk friction material, b) vehicle characteristics, such as vehicle weight and suspension type, c) maintenance history of the vehicle, d) driving behaviour, mainly associated with the speed, acceleration, and frequency and severity of braking events, e) road surface characteristics, and f) environmental conditions (Kukutschová et al., 2011; Kwak et al., 2013; Mosleh et al., 2004; Olofsson and Olander, 2013; Österle et al., 2001). Considering that braking events have a significant impact on the amount of material lost, the highest concentrations of brake and tyre wear particles should be observed near busy intersections, traffic lights, pedestrian crossings, corners and bus stops (e.g. Goel and Kumar, 2015; Limo et al., 2018). Several key factors need to be assessed when investigating wear particles toxicity and their potential harmful health effects, namely their size distribution, agglomeration state, chemical composition, surface area, chemistry and surface charge (Grigoratos and Martini, 2015 and references therein).

The Pb concentrations showed to be significantly higher in the car and bus comparing with the bicycle and metro samples ($p < 0.05$). The high concentrations of Pb in the road transport can be explained with the presence of Pb sources associated with brake and tyre wear and a small contribution from exhaust emissions as referred by Denier van der Gon and Appelman (2009). Despite the fact that leaded gasoline was banned in the European Union as of January of 2000, Pb is still found in gasoline as an impurity of crude oil (Pacyna et al., 2007). Thus, even if in trace amounts, gasoline is still an active and important source of Pb in Europe due to the huge amount of fuel used. Furthermore, it has been shown that resuspension of road dust contaminated with Pb usage in the past, is still a significant source of Pb in several urban areas (Lough et al., 2005; Young et al., 2002). The lower Pb concentrations found when commuting by bicycle (Fig. 4) may be due to the fact that the bike path passes through a garden in a section of the route, being away from the road traffic emissions.

The sea salt (mainly characterised by the presence of Na and Cl) found in the metro microenvironment and in-cabin of cars and buses is expected to come from outdoors. The concentrations of NaCl are very dependent on the meteorology and are affected by the close proximity to the Atlantic coast.

2.2. *Personal dose during commuting*

Several key factors need to be considered when investigating aerosol toxicity and potential negative health outcomes, with their chemical composition being one of the most important factors. The inhaled dose of air pollutants was calculated considering the time spent daily commuting and respective inhalation rates. The inhaled air volume increase from 2.4- to 3.5-fold times for active versus passive travel modes (considering both inhalation rates and journey times). Different patterns in inhaled pollutant dose were found among urban transport modes (Fig. 5).

The highest inhaled dose of PM2.5 was experienced when biking (55.0 μg), followed by metro (27.2 μg), bus (20.9 μg) and car (17.5 μg) travel. The same trend was observed for the inhaled dose of mineral elements, except for Fe where the highest dose was observed in the metro (7.6 μg; Fig. 5). Active travel has been frequently associated with higher doses of PM2.5, with cyclist-to-motorised travel dose ratios varying from about 1.2 to 2.6 in Guildford (UK) (Kumar et al., 2018), and with values around 1.2 in Barcelona (Spain) (de Nazelle et al., 2012). These results may be attributed not only to the higher travel durations but also to the higher physical effort in active travels. Relative to PM2.5, the mean inhaled dose for PM10 were around 1.1, 1.5 and 1.7 times higher during bicycle, bus and car modes, respectively, while for metro mode was 2.4. Thus, metro mode experienced the highest inhaled doses for PM10. The high difference in inhaled dose between PM10 and PM2.5 can be explained by the variations in PM mass concentrations. Although cars showed the lowest PM2.5 and PM10 inhaled doses among the transport modes studied, it should be emphasised that they have the highest emission per commuter. Therefore, the usage of private cars is actually the major cause of the increased exposure and dose of those commuters choosing an environmental-friendly transport mode.

The highest BC inhaled doses were also obtained when biking. Previous studies looking at the interaction of physical activity and BC exposure verified that the air pollution inhalation provoked lung function decreases while physical activity acted as an acute bronchodilator (illustrated by a significant increase in lung function) providing a potential protective effect (Laeremans et al., 2018; Matt et al., 2016).

On average, a metro traveller in Lisbon may inhale from 3.5 to 36.3 times more Fe during the daily commute, depending on the transport mode chosen to move around the city. This high value of Fe in metro was accompanied by the highest inhaled dose of Mn trace element, both elements associated to indoor sources as already discussed. In buses the inhaled doses of transition metals, particularly of Cu, Zn and Ni were from 2.4 to 37.6 times higher than in the other transport modes. In cars this behaviour only occurs for Sb (Fig. 5). In several epidemiological studies, metals such as Fe, Cu, Ni, Cr, Zn, Pb, S and Sb have been recognized as dangerous or potentially dangerous for the human health (e.g. Brook et al., 2010; Karlsson et al., 2005; Kelly and Fussell, 2012; Ostro et al., 2007; von Uexküll et al., 2005). High concentrations of these elements are measured in urban traffic environments; therefore, the health hazard from such particles should not be neglected.

The determination of the inhaled dose of aerosol chemical components by transport mode is a key factor to a comprehensive understanding of the possible health risks associated with such air pollutants. Moreover, the inhaled dose is driven

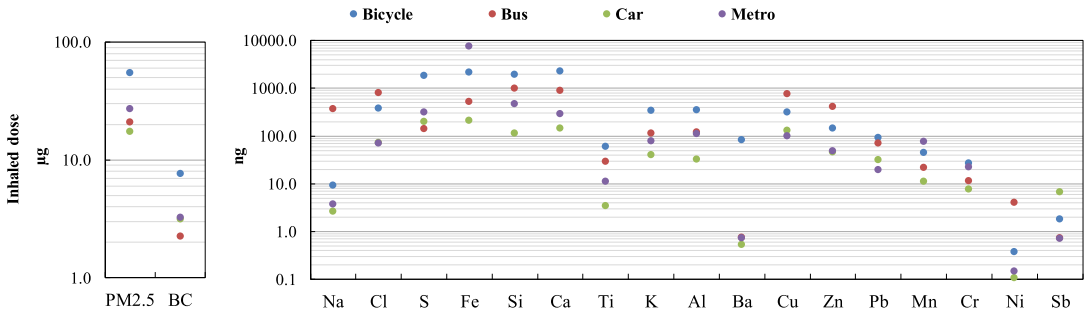

Fig. 5 – Inhaled dose of PM2.5 and its chemical components during a daily commuting in each transport mode.

by dynamic variables such as inhalation rates and pollutant concentrations among different transport modes and therefore, an accurate assessment of these two parameters is fundamental.

3. Conclusions

This study reports the personal exposure to PM and its chemical components using four different commuting modes, including bicycle, bus, car and metro in an urban environment. The main findings drawn from this study can be summarised as follows:

- The air pollutant concentrations that the urban commuters are exposed to vary significantly depending on the transport mode. The highest PM concentrations were generally observed when travelling by metro, but when inhalation rates and journey times were accounted for, the cyclists experienced the greatest inhaled doses.
- Cyclists' exposure to air pollutants are minimised by increasing distance from motorised traffic. Thus, the new urban planning policies should consider the development of alternative routes for cyclists away from busy roadways.
- PM2.5 mean concentrations in-vehicles (cars and buses) were lower than those in the respective ambient air, evidencing that the vehicle cabin may offer a protective effect. However, enhanced concentrations of certain trace elements, notably Zn and Cu, in-vehicles (I/O=22.3 and 10.7 for Zn in car and bus, respectively; and 15.1 and 8.9 for Cu in bus and car, respectively) were attributed to the presence of brake and tyre wear particles entering through windows, doors, air vents and other openings, since vehicles are not airtight.
- On average, BC concentrations when travelling by car were from 1.4 to 2.0 times higher than in the other transport modes, which was related to the closest proximity to the exhaust of other vehicles. This emphasises that BC concentrations serve as good proxy for road traffic exhaust emissions.
- PM chemical composition varied greatly depending on transport mode. In particular, Fe, Zn, Cu, Ni and K were considerably correlated in the motorised transport modes, indicative of a common traffic-related source, i.e. typical tracers of brake and tyre wear. In metro there was a dominant content of ferruginous particles, associated with the abrasion of rail-wheel-brake interfaces.
- In addition to their direct exposure to road traffic emissions, urban cyclists can inhale more particles of mineral origin, whereas metro commuters inhale a clear mixture of anthropogenic particles enhanced in Fe and Mn. The highest inhaled doses of Cu and Zn elements are observed when travelling by bus.

Emissions from vehicle exhausts have been decreasing through the development of cleaner technologies. Automotive industry is currently faced with the challenge of developing novel technologies for reducing non-exhaust emissions, without compromising performance and safety. The maintenance of the road surface should also be considered to minimize the emissions associated with road wear. This work highlights the increasing need for further research in this area. Moreover, the study results may help citizens to make more informed decisions when choosing a transport mode, in order to reduce their exposure to air pollutants.

Declaration of Competing Interest

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

Acknowledgements

This work was supported by European Union through the project LIFE Index-Air (LIFE15 ENV/PT/000674). This work reflects only the authors' view and EASME is not responsible for any use that may be made of the information it contains. This study was also supported by Portuguese Foundation for Science and Technology (FCT) through the projects ExpoLIS (LISBOA-01-0145-FEDER-032088) and UID/Multi/04349/2013, the contract CEECIND/04228/2018 and the PhD grants SFRH/BD/129149/2017 and SFRH/BD/147074/2019. The authors also gratefully acknowledge the permission and collaboration from the public transportation company of Lisbon CARRIS to undertake this work.

Appendix A Supplementary data

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.jes.2020.07.008.

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