



## Urban air quality comparison for bus, tram, subway and pedestrian commutes in Barcelona



Teresa Moreno <sup>a,\*</sup>, Cristina Reche <sup>a</sup>, Ioar Rivas <sup>a</sup>, María Cruz Minguillón <sup>a</sup>, Vânia Martins <sup>a</sup>, Concepción Vargas <sup>b</sup>, Giorgio Buonanno <sup>b,c</sup>, Jesús Parga <sup>a</sup>, Marco Pandolfi <sup>a</sup>, Mariola Brines <sup>a</sup>, Marina Ealo <sup>a</sup>, Ana Sofia Fonseca <sup>a</sup>, Fulvio Amato <sup>a</sup>, Garay Sosa <sup>a</sup>, Marta Capdevila <sup>d</sup>, Eladio de Miguel <sup>d</sup>, Xavier Querol <sup>a</sup>, Wes Gibbons <sup>e</sup>

<sup>a</sup> Institute of Environmental Assessment and Water Research (IDAE-CSIC), C/Jordi Girona 18-24, 08034 Barcelona, Spain

<sup>b</sup> DICeM—University of Cassino and Southern Lazio, via Di Biasio 43, 03043 Cassino, FR, Italy

<sup>c</sup> International Laboratory for Air Quality and Health, Queensland University of Technology, Brisbane, Qld, Australia

<sup>d</sup> Transports Metropolitans de Barcelona, Santa Eulalia, Av. del Metro s/n, 08902 L'Hospitalet de Llobregat, Barcelona Spain

<sup>e</sup> WPS, C/Major 13, 08870 Sitges, Spain

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### ABSTRACT

Access to detailed comparisons in air quality variations encountered when commuting through a city offers the urban traveller more informed choice on how to minimise personal exposure to inhalable pollutants. In this study we report on an experiment designed to compare atmospheric contaminants inhaled during bus, subway train, tram and walking journeys through the city of Barcelona. Average number concentrations of particles 10–300 nm in size,  $N$ , are lowest in the commute using subway trains ( $N < 2.5 \times 10^4 \text{ part. cm}^{-3}$ ), higher during tram travel and suburban walking ( $2.5 \times 10^4 \text{ cm}^{-3} < N < 5.0 \times 10^4 \text{ cm}^{-3}$ ), and highest in diesel bus or walking in the city centre ( $N > 5.0 \times 10^4 \text{ cm}^{-3}$ ), with extreme transient peaks at busy traffic crossings commonly exceeding  $1.0 \times 10^5 \text{ cm}^{-3}$  and accompanied by peaks in Black Carbon and CO. Subway particles are coarser (mode 90 nm) than in buses, trams or outdoors (< 70 nm), and concentrations of fine particulate matter (PM<sub>2.5</sub>) and Black Carbon are lower in the tram when compared to both bus and subway. CO<sub>2</sub> levels in public transport reflect passenger numbers, more than tripling from outdoor levels to > 1200 ppm in crowded buses and trains. There are also striking differences in inhalable particle chemistry depending on the route chosen, ranging from aluminosilicate at roadsides and near pavement works, ferruginous with enhanced Mn, Co, Zn, Sr and Ba in the subway environment, and higher levels of Sb and Cu inside the bus. We graphically display such chemical variations using a ternary diagram to emphasise how “air quality” in the city involves a consideration of both physical and chemical parameters, and is not simply a question of measuring particle number or mass.

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### 1. Introduction

Most modern cities have serious environmental issues linked to pervasive airborne particle emissions from road traffic and other anthropogenic sources. The resulting effect on urban human

health depends on a complex mix of factors that includes airborne particle concentration, size, and composition, the distribution of pollutants in time and space, and the physical condition of the urban traveller. Given this inherent complexity, and a corresponding lack of consensus among researchers as to which PM characteristics most closely relate to health effects, the simple act of measuring average pollutant concentrations at background monitoring stations or traffic hot-spots (especially in the case of ultrafine particles) offers only a general guide to actual variations in airborne particle concentrations and compositions inhaled by people moving through the city (Kaur et al., 2007; Zuurbier et al., 2010; de Nazelle et al., 2012). This is of particular relevance to regular commuters, who will receive a widely differing dose of air

**Abbreviations:** PM<sub>2.5</sub>, particulate matter with a diameter below 2.5 microns; PEM, Personal Environmental Monitor; ICP-AES, Inductively Coupled Plasma Atomic Emission Spectroscopy; ICP-MS, Inductively Coupled Plasma Mass Spectrometry; CPC, condensation particle counter;  $N$ , particle number concentrations; BC, Black Carbon; SCRT<sup>+</sup>, Selective Catalytic Reduction+Continuously Regenerating Trap; ADSA, alveolar deposited surface area

\* Corresponding author. Fax: +34 934110012.

E-mail address: [teresa.moreno@idaea.csic.es](mailto:teresa.moreno@idaea.csic.es) (T. Moreno).

pollutants depending on the mode of transport/microenvironment they select to get to and from work and the area of the city where they commute.

Previous publications have identified many of the influences on the distribution of transport-related air pollutants in the city environment (e.g. Adams et al., 2001; Hammond et al., 2006; Kaur et al., 2006, 2007; Kaur and Nieuwenhuijsen, 2009; Moreno et al., 2009; Zuurbier et al., 2010; Knibbs et al., 2011; de Nazelle et al., 2012; Kingham et al., 2013; Karanasiou et al., 2014; Gu et al., 2015; Liu et al., 2015). Levels of particulate mass will depend on such factors as traffic intensity (Zhu et al., 2002; Buonanno et al., 2011), time of the day (Kaur et al., 2007; Dons et al., 2011; de Nazelle et al., 2012), meteorology (Gomez-Perales et al., 2004; Kaur et al., 2007; Buonanno et al., 2011), type of vehicles (Dons et al., 2011), and distance to the road (Kaur et al., 2006, 2007). In terms of particle number ( $N$ ), most inhalable traffic particles fall into the ultrafine size range (particles with a diameter less than 100 nm) (Morawska et al., 2008), with diesel buses providing an especially potent pollutant source (Kaur et al., 2007; Knibbs and de Dear, 2010; Kingham et al., 2013) and comparing unfavourably with, for example, gas- or electric powered buses (Zuurbier et al., 2010; Dons et al., 2011; Ragettli et al., 2013). Indoor controls such as the use of strong ventilation in subway platforms (Querol et al., 2012) or air conditioning in underground trains can have a clearly beneficial effect (Martins et al., 2015a). In addition to Particulate Matter (PM) distribution, CO levels provide another marker for vehicle exhaust emissions in the urban environment, with typically higher concentrations breathed when travelling by car (Kingham et al., 2013) or bus (Kaur et al., 2007; de Nazelle et al., 2012) compared to walking.

In this paper we offer further insight into the reality of variations in urban air quality experienced during travel on different forms of public transport, and compare these to conditions while walking in the city. Our study uses continuously measuring portable equipments carried by two commuters making journeys through the city with the same start and end point and at the same time but using different transport modes (bus, subway, tram and walking). The study continuously tracks and compares not only PM mass and  $N$  during each journey, but also Black Carbon,

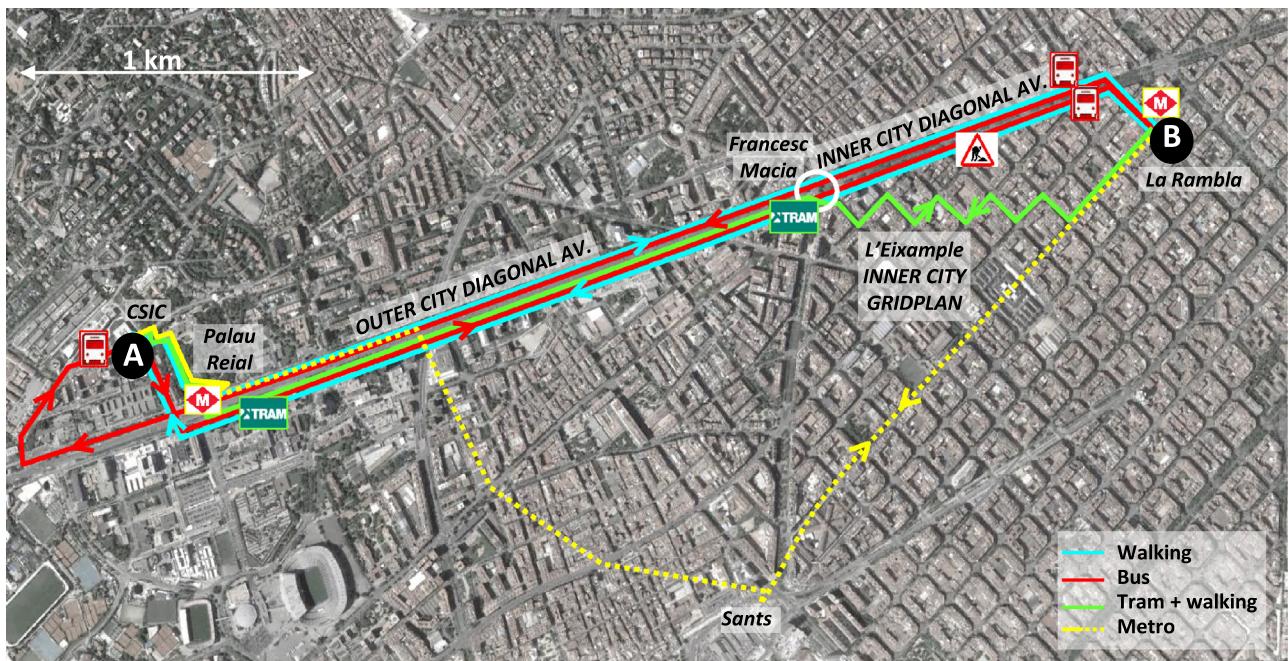
Carbon Monoxide, Carbon Dioxide and, as far as we are aware, for the first time includes chemical composition of the finer material inhaled ( $PM_{2.5}$ ) by individuals whilst moving through the city.

## 2. Methodology and working conditions

The monitoring equipment was carried in backpacks by two commuters during October and November 2014, each separately and simultaneously making a round trip route (carrying a GPS) using one of the four selected transport modes in the city of Barcelona for a total of 39 weekdays, with only one day with rain. The commuter pairs began their journey together but took different routes through the city. The commute chosen was 8.4–9 km long (4.2–4.5 km each way depending on the transport mode) from the suburban area of the IDAEA-CSIC Institute in Pedralbes to the Diagonal metro stop on La Rambla in the city centre (Fig. 1). Four types of commute were included in the study: walking only, walking+tram, bus, and subway train (Metro).

**Walking only:** The pedestrian-only route followed the sidewalks of the Diagonal Avenue, a straight multilane urban highway that provides a key arterial entry and exit route for the city. Despite the heavy traffic along this highway (132,000 vehicles per day), it is used by many pedestrians and cyclists. There is a clear division between the suburban and city centre sectors of this highway, demarcated around halfway into the journey by the Francesc Macia roundabout (Fig. 1). Whereas the suburban sector (west side) is open and with generally wide sidewalks (100 m wide), the city centre sector is more severely congested, more narrow (45 m) and canyon-like and with a higher number of crossroads. At the time of our monitoring experiment a major restructuring of this central part of the Diagonal was taking place, involving extensive repaving and associated uncontrolled emissions under dry conditions, providing us with an interesting opportunity to highlight the effect of such works (which are not uncommon in the city) on air quality. The same route was followed each day, with the outward journey along the northern side of the Diagonal, and the return journey being made on the opposite side.

**Walking+tram:** Because of the special conditions produced by



**Fig. 1.** Selected routes for measuring air quality while walking and travelling by bus, tram and metro. The figure shows the locations of the beginning (A, CSIC) and final (B, La Rambla) point of the route, the tram, bus and metro stops and the area with important street works.

the extensive works along the central Diagonal, an alternative walking route from Francesc Macia to the centre was selected. This was made using the distinctively chamfered perpendicular grid pattern 19th century enlargement of the city known as L'Eixample, doglegging alternately right and left along 13 m-wide streets to reach La Rambla (Fig. 1). As with the previously described journey, the same route was followed each day, using opposite sides of the sidewalk for outward and return journeys. The journey to the Francesc Macia roundabout in this case was made using the tram running down the suburban sector of the Diagonal from Palau Reial (Fig. 1); at present the modern and expanding Barcelona tram system does not penetrate the city centre.

**Bus:** The bus trip was done taking Route 33 (articulated diesel bus, 18 m long, all Euro -2 and Euro-3 equipped with SCRT<sup>®</sup>) from the IDAEA-CSIC entrance, and involved 350 m of walking to La Rambla at the journey's end (Fig. 1). When entering the bus the traveller was always in the central part of the vehicle, away from any of the doors.

**Metro:** The underground rail route involved travelling initially using Line 3 from Palau Reial and changing in Sants station to Line 5, thus including two trains each way, waiting on 2 different platforms and walking in the long connecting subway corridor to make the change.

A total of 78 trips were monitored, with a return route that lasted between 80 (bus and metro) to 120 min (walking and tram). The journeys started at 10:00 am avoiding the height of the rush hour (although the city is by then extremely busy), and notes were taken by each commuter to record the timing, location, progress and any special events during each journey (road crossing, bus, tram and metro stops, roadworks, getting in and out of the vehicle, etc.).

PM<sub>2.5</sub> gravimetric samples were collected in 63 mm quartz microfiber filters using a Personal Environmental Monitor (PEM) with a flow rate of 10 L/min and chemically analysed using ICP-AES and ICP-MS for major and trace elements respectively. SiO<sub>2</sub> and CO<sub>3</sub><sup>2-</sup> were indirectly determined on the basis of empirical factors (Al × 1.89 = Al<sub>2</sub>O<sub>3</sub>, 3 × Al<sub>2</sub>O<sub>3</sub> = SiO<sub>2</sub> and (1.5 × Ca) + (2.5 × Mg) = CO<sub>3</sub><sup>2-</sup>, see Querol et al., 2001). Each transport mode was repeated until enough PM sample had been obtained for chemical analysis, thus each subject carried the same filter sample during 5–7 days using the same transport mode. Therefore we analysed 6 filters per subject, this is 12 filters in total including 3 for each of the four transport modes. PM<sub>2.5</sub> concentrations were measured using a DustTrak monitor (DT: Model 8533, TSI) and corrected based on a comparison against simultaneous gravimetric PM<sub>2.5</sub> (MCV High Volume sampler) at the urban background site of Palau Reial in Barcelona after the sampling campaigns. Although a good relative correlation was observed between PM<sub>2.5</sub> urban background measures in the local station and the values registered in the DustTrak while walking ( $R^2=0.82$ ) the latter were always much higher in absolute values. The correlation between the PM<sub>2.5</sub> concentrations determined by the gravimetric PM<sub>2.5</sub> concentrations from PEM filters and the DustTrak showed a poorer correlation ( $R^2=0.55$ ) and again DustTrak values show much higher concentrations. This we attribute to the portability of the equipment because DustTrak values obtained are more reliable when the equipment is quietly measuring on a flat surface, but less optimal while being carried in a rucksack. In any case a tendency towards overestimation of PM mass values measured by DustTrak has already been demonstrated by previous works (Knibbs and de Dear, 2010), although uncorrected values can usefully show relative concentrations between different microenvironments, which is how our data are treated here rather than applying reference correction factors (cf. Quiros et al., 2013).

The N concentrations in the range of 10–300 nm were measured using a NanoTracer (Philips Aerasense Nanotracer, NT)

which can also estimate the different fractions of lung deposited surface area through a semi-empirical algorithm implemented by Marra et al. (2010). The NT was operated in Advanced mode, wearing it around the waist using a dedicated belt. A particle number concentration correction factor was derived for each NT at the start of the project by running side by side with a TSI model 3775 condensation particle counter (CPC) calibrated at the European Accredited Laboratory of the University of Cassino and Southern Lazio (Italy). The particle surface area concentration per unit volume of inhaled air that is likely to be deposited in various regions of the respiratory tract can be also estimated through the NanoTracer (Buonanno et al., 2014a). This is a significant aspect since deposited surface area concentration is currently of great attention to the scientific community, for use as an important metric for assessing the relative exposure-induced health risk for a given particle hazard (Giechaskiel et al., 2009; Cauda et al., 2012). In particular, the alveolar deposited surface area concentration, SAL (namely Fuchs surface area, Jung and Kittelson, 2005; it is expressed in  $\mu\text{m}^2 \text{cm}^{-3}$ ), is calculated by the NanoTracer according to the following equation:

$$S_{\text{AL}} \approx 5.4 \times 10^{-2} \cdot N \cdot d_{p,\text{av}} \propto I_1 \quad (1)$$

where the total particle number, N, is expressed in  $\text{part. cm}^{-3}$ , the number-averaged particle size, dp, av, in  $\mu\text{m}$  and  $I_1$  is the total particle charge deposited per unit time inside the Faraday cage (Marra et al., 2010).

Black Carbon (BC) was measured using a mini-aethalometer (MicroAeth AE51) and carbon dioxide and monoxide with a IAQ-Calc equipment (Model 7545, TSI) which also recorded T and RH values. Further details on monitoring, analytical procedures and meteorological conditions are provided in Supplementary Information and Fig. S1.

Time resolution for the continuous measurements were made according to the capabilities of each equipment, varying from 16 s (PMx and N), 20 s (CO, CO<sub>2</sub>) to 30 s for BC. Values registered while commuting by walking were sub-divided into those while walking in the suburban part of the Diagonal west of Francesc Macia (*outer Diagonal*), those registered while walking in the Diagonal Avenue in the second part (with important pavement street works, *inner Diagonal*), walking in the L'Eixample district after leaving the tram (with narrower and high traffic streets, *inner gridplan*) and walking through La Rambla (after leaving the bus) (Fig. 1).

A normality test was applied to determine the type of distribution of the data, if parametric (normal) or non-parametric. The distribution of the N concentration was found to be not symmetric but skewed-right, due to few high values increasing the mean value, but this does not affect the median. Therefore, the Kruskal-Wallis test was applied for comparing two or more samples that are independent, based on analysis of variance: a p-value < 0.01 was regarded as statistically significant.

### 3. Results

#### 3.1. Number and size of ultrafine particles

Overall average N concentrations and size modes of ultrafine particles are compared in Table 1. Data were separated into sub-modes involving only tram, bus, metro or walking in different environments (Fig. 1). In terms of public transport used, N is lowest in the metro ( $2.3 \times 10^4 \text{ cm}^{-3}$ ), higher in the tram ( $3.0 \times 10^4 \text{ cm}^{-3}$ ) which runs only through the outer Diagonal, and highest in the bus ( $5.4 \times 10^4 \text{ cm}^{-3}$ ). With regard to particle size, the reverse hierarchy is the case, with the highest mode values occurring in the metro (90 nm instead of 64–66 nm). In this table

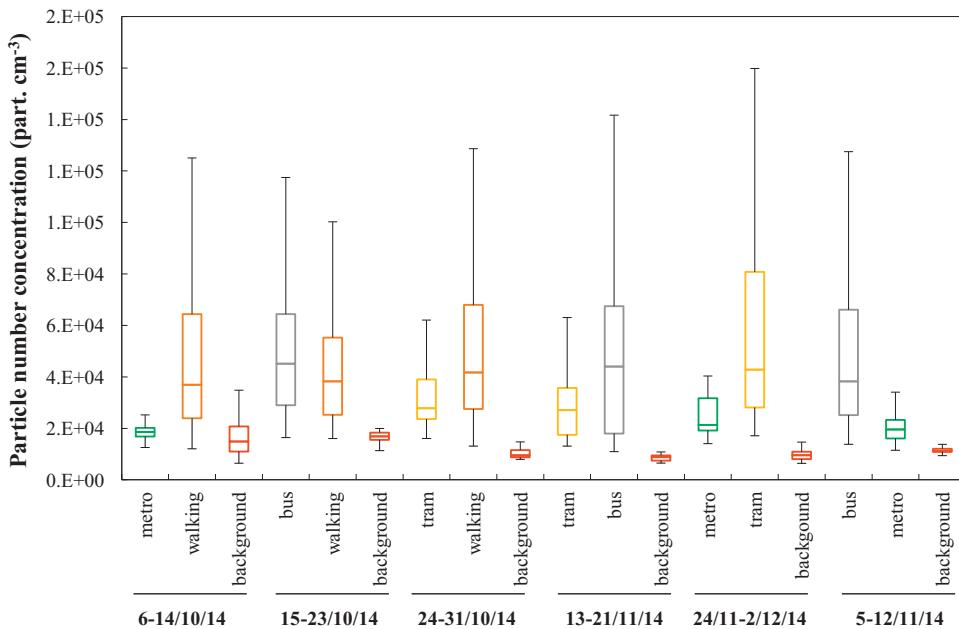
**Table 1**

Arithmetic mean and median concentrations and standard deviation values (in italics) for the particle number, size mode, BC, CO<sub>2</sub> and CO for each type of transport. \*Possibly overestimated by interference of light absorption by Fe in the microethalometre measurements.

Metro			Tram			Bus		
Mean	St. dev	Median	Mean	St. dev	Median	Mean	St. dev	Median
<b>N (part. cm<sup>-3</sup>)</b>	$2.3 \times 10^4$	$0.4 \times 10^4$	$2.1 \times 10^4$	$3 \times 10^4$	$1 \times 10^4$	$2.8 \times 10^4$	$5.4 \times 10^4$	$1.6 \times 10^4$
<b>Mode (nm)</b>	90	13	91	66	9	67	64	8
<b>BC (µg m<sup>-3</sup>)</b>	$7.0^*$	3.5	6.5	3.4	2.2	3.0	5.5	3.6
<b>CO<sub>2</sub> (ppm)</b>	694	63	631	643	80	660	721	158
<b>CO (ppm)</b>	0.9	0.4	0.9	0.4	0.3	0.5	0.9	0.3

Walking											
Outer diagonal			Inner diagonal			Central gridplan			Diagonal to La Rambla		
Mean	St. dev	Median	Mean	St. dev	Median	Mean	St. dev	Median	Mean	St. dev	Median
<b>N (part. cm<sup>-3</sup>)</b>	$3.7 \times 10^4$	$0.6 \times 10^4$	$3 \times 10^4$	$5.9 \times 10^4$	$1.3 \times 10^4$	$4.8 \times 10^4$	$5.4 \times 10^4$	$1.8 \times 10^4$	$4.4 \times 10^4$	$5.4 \times 10^4$	$2 \times 10^4$
<b>Mode (nm)</b>	66	9	63	56	9	55	54	6	53	63	7
<b>BC (µg m<sup>-3</sup>)</b>	6.5	2.4	5.2	9.6	2.7	7.9	5.3	3.0	4.1	4.4	2.9
<b>CO<sub>2</sub> (ppm)</b>	456	20	450	464	21	463	425	44	406	479	68
<b>CO (ppm)</b>	0.8	0.2	0.7	1.1	0.2	1.1	0.7	0.3	0.8	0.9	0.3

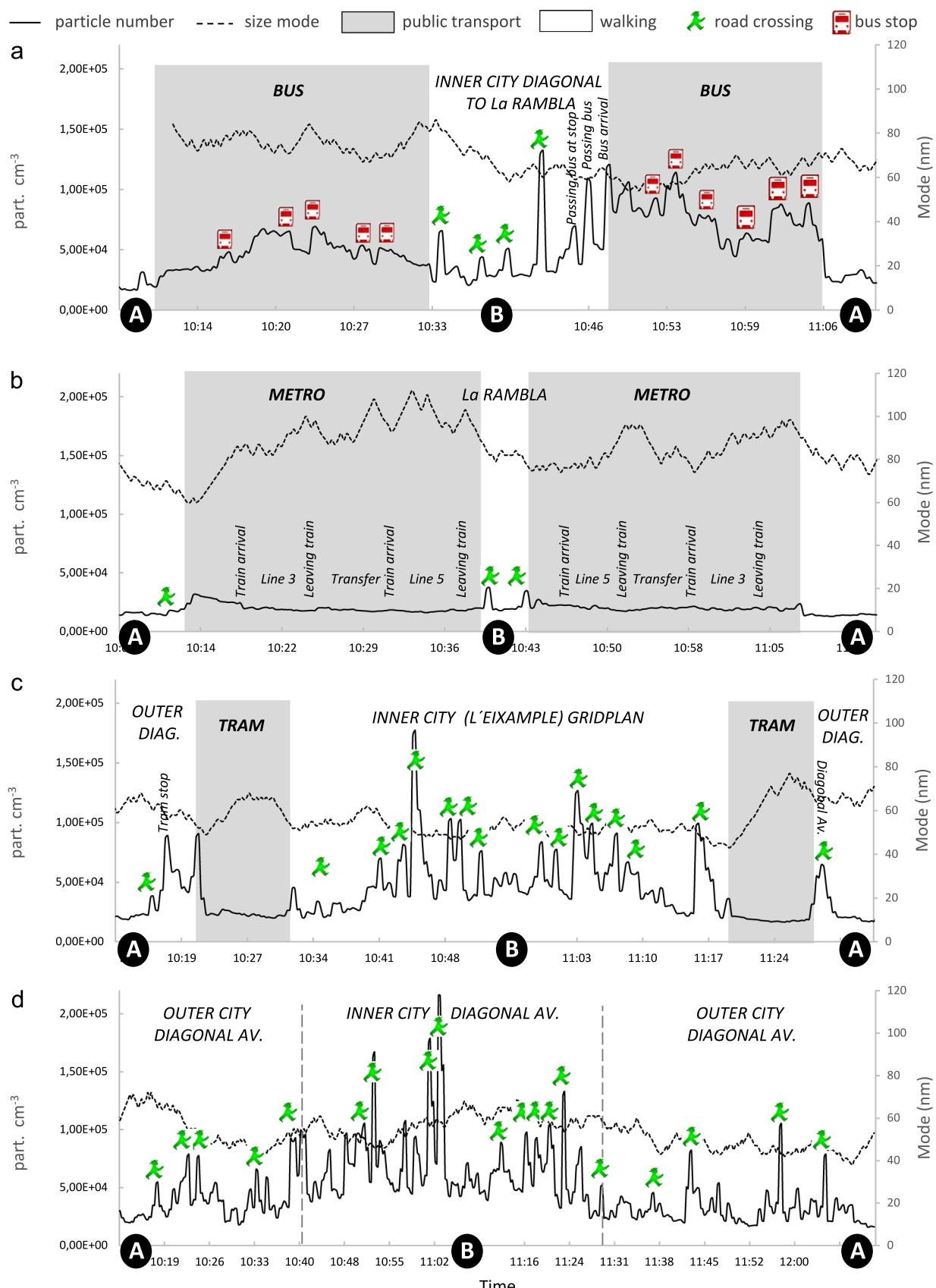


**Fig. 2.** Statistics of the particle number concentration comparing bus, tram, metro and pedestrian commutes to the background measured during the same day.

we also compare the various pedestrian segments of the journey when the commuter was walking through the suburban area west of Francesc Macia (outer Diagonal,  $3.7 \times 10^4 \text{ cm}^{-3}$ ), the inner city L'Eixample gridplan ( $5.4 \times 10^4 \text{ cm}^{-3}$ ) and inner central Diagonal ( $5.9 \times 10^4 \text{ cm}^{-3}$ ). All these values were much higher than those registered in the urban background site of Barcelona where annual average particle number below 1 µm for 2014 was  $1.4 \times 10^4 \text{ cm}^{-3}$  (this being measured with a water-based CPC). Using median values, N is again lowest in the metro ( $5.0 \times 10^3 \text{ cm}^{-3}$ , equal to 31% of the background value), higher in the tram ( $1.1 \times 10^4 \text{ cm}^{-3}$ , 63%) and highest in the bus ( $2.8 \times 10^4 \text{ part. cm}^{-3}$ , 154%). The pedestrian journey shows an overall particle number concentration increase equal to  $1.9 \times 10^4 \text{ cm}^{-3}$  (112%). Details of the statistics for each commuter trip are reported in Fig. 2.

Fig. 3 shows N variability comparing continuous real-time measurements of N and their size modes on each of the four commuting return journeys. With regard to the journey using buses, because this begins in a quiet suburban setting just one stop

after the beginning of the bus route, N initially presents a low value ( $< 3.0 \times 10^4 \text{ cm}^{-3}$ ) but increases as the vehicle travels along the Diagonal, with minor peaks resulting from doors opening at bus stops (Fig. 3a). Upon leaving the bus, background conditions on the sidewalk are better than in the bus, although punctuated by high-amplitude transient peaks at road crossings, especially when crossing the Diagonal or waiting at the roadside for the return bus. The N concentration (mode about 60 nm) during these roadside peak events not uncommonly exceeds  $1.0 \times 10^5 \text{ cm}^{-3}$  (Fig. 3a). During the return bus, initial indoor air quality in terms of N is notably worse than on the outward journey as a consequence of the bus having travelled through the congested city centre, although it does improve somewhat once the bus passes the most congested parts of the Diagonal avenue. It is also noticeable how the size mode reduces during the return journey, presumably reflecting increased contamination by fresh traffic fumes entering the bus (see discussion below of the CO data which demonstrate higher fresh traffic emissions on the northern, downwind, side of



**Fig. 3.** Distribution of particle number concentrations and size mode during the route done by bus (a), metro (b), tram + walking (c) and walking (d). The time spent inside public transport is shown in grey. (A: start point at CSIC, B: final point at La Rambla).

**Table 2**

Filter samples collected in parallel using four different commuting modes, indicating sampling dates and PM<sub>2.5</sub> mass concentrations ( $\mu\text{g m}^{-3}$ ). PM<sub>2.5</sub> column on the right shows the highest concentration. UB: urban background site in Palau Reial.

Time period	Transport mode	PM <sub>2.5</sub>	Transport mode	PM <sub>2.5</sub>	UB PM <sub>2.5</sub>
06–14/10/14	Walking	23	Metro	37	12
15–23/10/14	Walking	29	Bus	48	12
24–31/10/14	Walking	32	Tram+walking	35	17
13–21/11/14	Tram+walking	27	Metro	49	12
24/11–2/12/14	Metro	42	Bus	49	11
05–12/11/14	Tram+walking	29	Bus	39	10

the outer Diagonal).

The marked difference in terms of N between travelling by bus and in the metro train is demonstrated by comparing Fig. 3a and b. Notable transient peaks experienced during the metro journey are few and related to exposure to traffic fumes on entry and exit from the underground system. There are slight increases during train arrival, but compared to conditions above ground fine/ultrafine N are remarkably stable and relatively low on platforms, inside trains and when walking through station transfer tunnels. Equally clear is the higher value of the N mode at underground when compared with the more traffic-contaminated environment above ground.

N concentrations measured within the interior of the tram are very stable and much lower than in the bus (compare Fig. 3a and c), despite the fact that the tram also runs alongside the Diagonal highway. This stability is lost however when the commuter leaves the tram for the traffic-congested walk through the inner city gridplan (Fig. 3c). As before, in this outdoor environment the most obvious feature is the number of high amplitude particle number transient peaks induced by passing and idling traffic, especially at road junctions. The characteristic highly irregular serrated patterns produced by these abundant peaks are accompanied by a drop in size mode to  $< 60 \text{ nm}$ . Although the most common source of the transient peaks is obviously traffic, some can be attributed at least in part to other sources such as passing smokers or bar terraces with butane gas heating. A similar pattern, although even more accentuated, is revealed by the data from the walking-only route down the Diagonal (Fig. 3d). At some road crossings in the highly polluted inner city part of this highway, N can briefly exceed  $2.0 \times 10^5 \text{ part.cm}^{-3}$  (Fig. 3d). When comparing the pairs of routes completed on the same days, through the application of the Kruskal–Willis test, the N concentration measured in the metro presents statistically significant differences with respect to the other forms of commuting considered. In contrast, we found few differences among walking, bus and tram commutes. Therefore, the particle number concentrations measured were significantly lowest in the commute using subway trains, emphasising the importance of road traffic as a major source of ultrafine particles.

### 3.2. Alveolar deposited surface area concentrations

Alveolar deposited surface area (ADSA) concentrations were estimated through the continuous data recorded every 16 s by the automatic NanoTracer monitors for every trip. The lowest average values where measured in the tram ( $92 \pm 27 \mu\text{m}^2 \text{cm}^{-3}$ ), with concentrations increasing through the metro ( $94 \pm 21 \mu\text{m}^2 \text{cm}^{-3}$ ), walking ( $101 \pm 20 \mu\text{m}^2 \text{cm}^{-3}$ ), to  $125 \pm 39 \mu\text{m}^2 \text{cm}^{-3}$  when travelling by bus. Therefore, the commuter using subway trains experiences the minimum particle concentrations in terms of N but in terms of ADSA it tends to increase relative to the other transport modes. In fact, the size mode of the particles encountered underground is larger than in outdoor environments freshly contaminated by traffic. Consequently, when considering the ADSA

concentration, the apparent advantage of metro over outdoor travel becomes less evident.

With respect to the comparison of the pairs of routes completed on the same days, the application of the Kruskal–Willis test to the ADSA concentration measured in the tram presents the greatest statistically significant differences with respect to the other forms of commuting considered, with little difference among walking, bus and metro commutes, but being significantly lower in the tram commute.

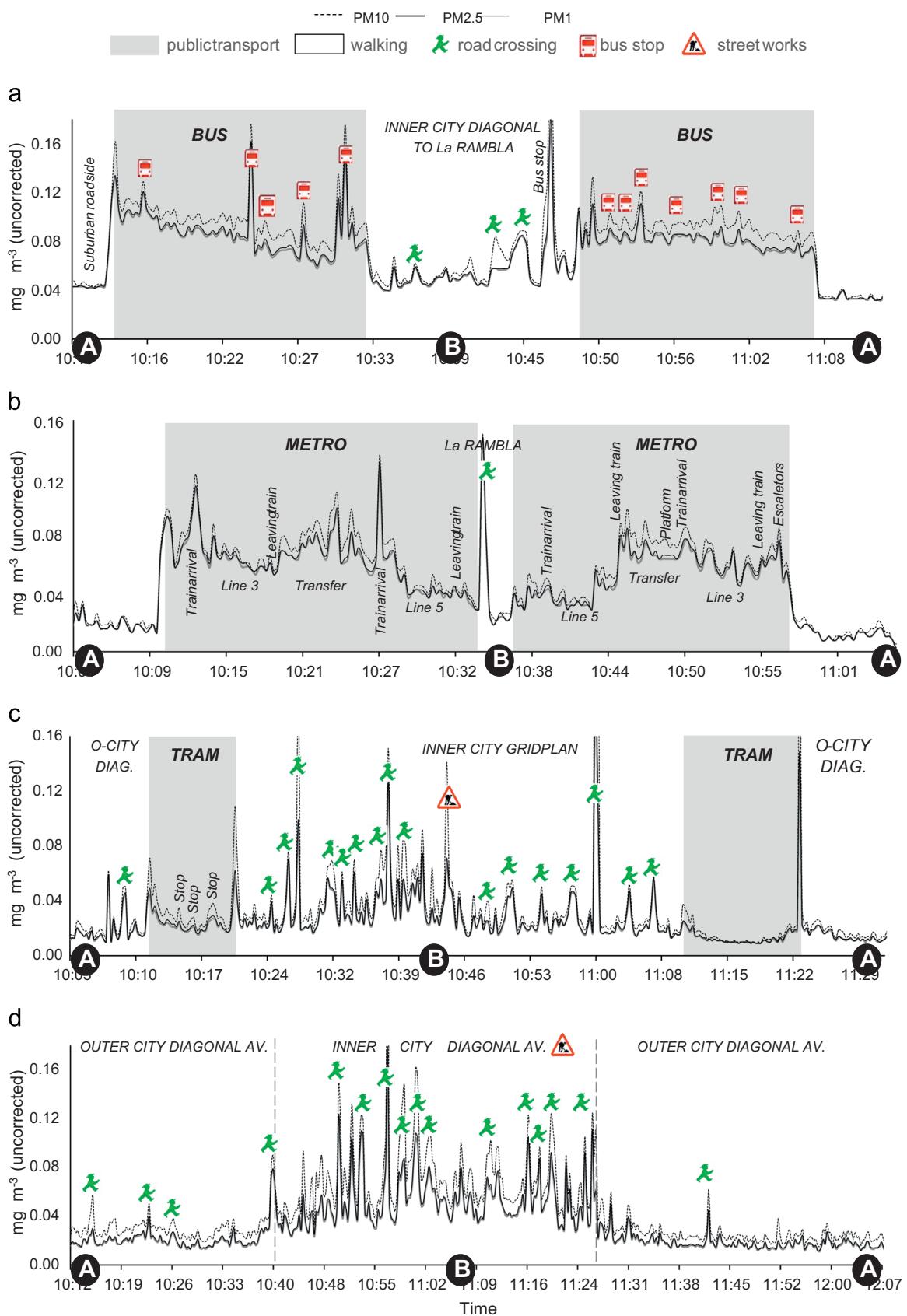
### 3.3. PM<sub>2.5</sub> concentrations

PM<sub>2.5</sub> mass concentrations were obtained from both the quartz fibre filters (each collected over 5–7 consecutive days) and the continuous data recorded by the automatic DustTrak monitor for every trip. The gravimetric results from the filters are presented in Table 2 and record lowest average values when walking ( $28 \mu\text{g m}^{-3}$ : range  $23$ – $32 \mu\text{g m}^{-3}$ ), with concentrations increasing through the tram+walking journey ( $30 \mu\text{g m}^{-3}$ : range  $27$ – $35 \mu\text{g m}^{-3}$ ), the metro ( $43 \mu\text{g m}^{-3}$ : range  $37$ – $49 \mu\text{g m}^{-3}$ ), to  $45 \mu\text{g m}^{-3}$  (range  $39$ – $49 \mu\text{g m}^{-3}$ ) when travelling by bus. The greatest variability between samples was observed when travelling in the metro, and the lowest when using the tram+walking route. When comparing the pairs of routes completed on the same days the walking route was always the one with lower PM<sub>2.5</sub> concentrations (Table 2).

The automatic uncorrected PM data from the DustTrak were separated, as in the case of N concentrations, into sub-modes involving only tram or walking in different environments (Fig. 1). When separating them, relative average PM<sub>2.5</sub> concentrations were lowest while travelling in the tram, and highest (56% higher) inside the bus. Of all walking routes, PM<sub>2.5</sub> relative mass concentrations were at their lowest in the outer Diagonal (still 11% higher than being inside the tram), and progressively increased while walking in the L'Eixample gridplan (30% higher than the tram), or in the inner Diagonal with extensive paving works (35% higher).

Fig. 4 compares the typical patterns of inhalable PM concentrations recorded during each of the four commuting journeys. Both bus and metro show a clear increase in inhalable particle mass inside compared to outdoor urban background (Fig. 4a, b). As with the particle number concentration data, transient peaks are again commonly encountered at stops during the bus journey and outside when walking across road junctions. In the metro the highest peaks are commonly associated with the effect of train arrival. Interestingly, there is a clearly detectable difference in air quality between the two rail journeys underground, with Line 3 recording inhalable PM mass concentrations over 50% higher than Line 5 (Fig. 4b). Another observation underground is that the air breathed during the transfer between lines has more PM<sub>10</sub> particles than on the platforms or inside trains (Fig. 4b). Fig. 4 demonstrates how, in terms of mass concentrations, tram travel has by far the best air quality of the three modes of public transport used, with only minor transient peaks produced at some of the busier stops (outward journey on Fig. 4c). With regard to the pedestrian segments of the journeys, the PM data record the same sequence of transient peaks associated with road junctions. Such peaks are highly variable depending on the aerosol micro-environment briefly enveloping the commuter, and in some cases are quite dramatic, as demonstrated by the peak registered on leaving the return tram to re-enter direct exposure to the Diagonal urban motorway plume (final peak on Fig. 4c).

In the city centre the influence of coarser particle resuspension and pavement works is emphasised by several peaks in which the PM<sub>10</sub> component shows an exceptional increase (e.g. the road-works labelled on Fig. 4c and several similar peaks on Fig. 4d). In



**Fig. 4.** Distribution of particulate matter mass concentrations (PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1</sub>) during the route done by bus (a), metro (b), tram + walking (c) and walking (d). The time spent inside public transport is shown in grey. Values are uncorrected and for relative comparison (A: start point at CSIC, B: final point at La Rambla).

the L'Eixample gridplan streets it was noted that PM concentrations are slightly higher when walking alongside traffic climbing away from the sea (e.g. Casanova and Aribau streets) as opposed to descending traffic (Villaroel and Muntaner streets), and higher when walking on the north side of the street (presumably related to the effect of the diurnal sea breeze driving airflow away from the coast). Outside the city centre, in the more suburban segment of the Diagonal, PM concentrations are lower, although again punctuated by prominent peaks at major crossings such as Gran Via Carles III, Numància and Av. Sarrià.

### 3.4. Black carbon (BC)

Average BC values were highest when walking in the congested inner city Diagonal ( $9.6 \mu\text{g m}^{-3}$ ), lower in the metro ( $7 \mu\text{g m}^{-3}$ ), when walking along the more open suburban Diagonal ( $6.5 \mu\text{g m}^{-3}$ ) and riding inside the bus ( $5.5 \mu\text{g m}^{-3}$ ), and lowest when travelling by tram ( $3.4 \mu\text{g m}^{-3}$ ). In all these cases concentrations were two to five times higher than the average urban background value of  $1.6 \mu\text{g m}^{-3}$  measured in Palau Reial for the year 2014 (Table 1).

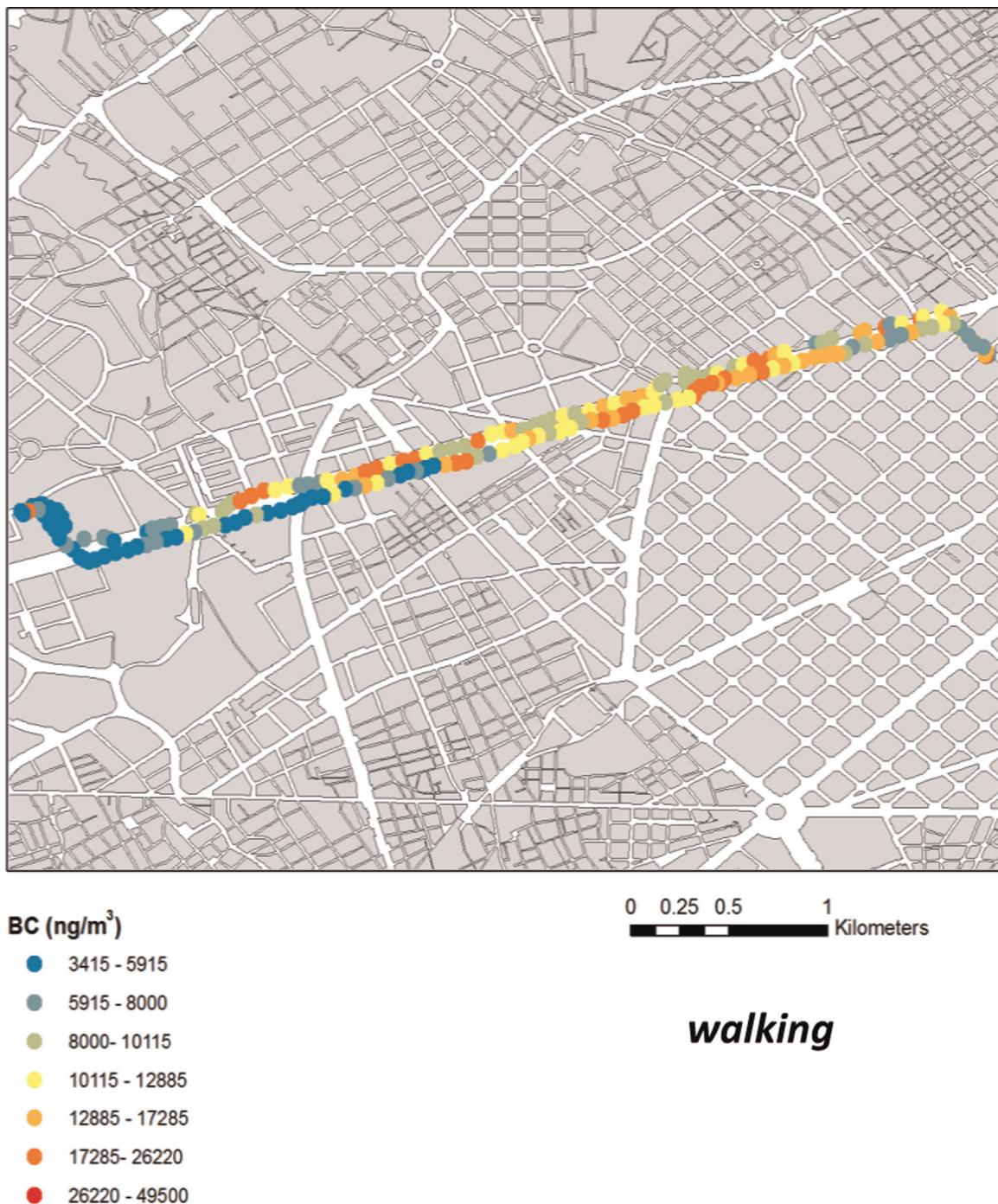
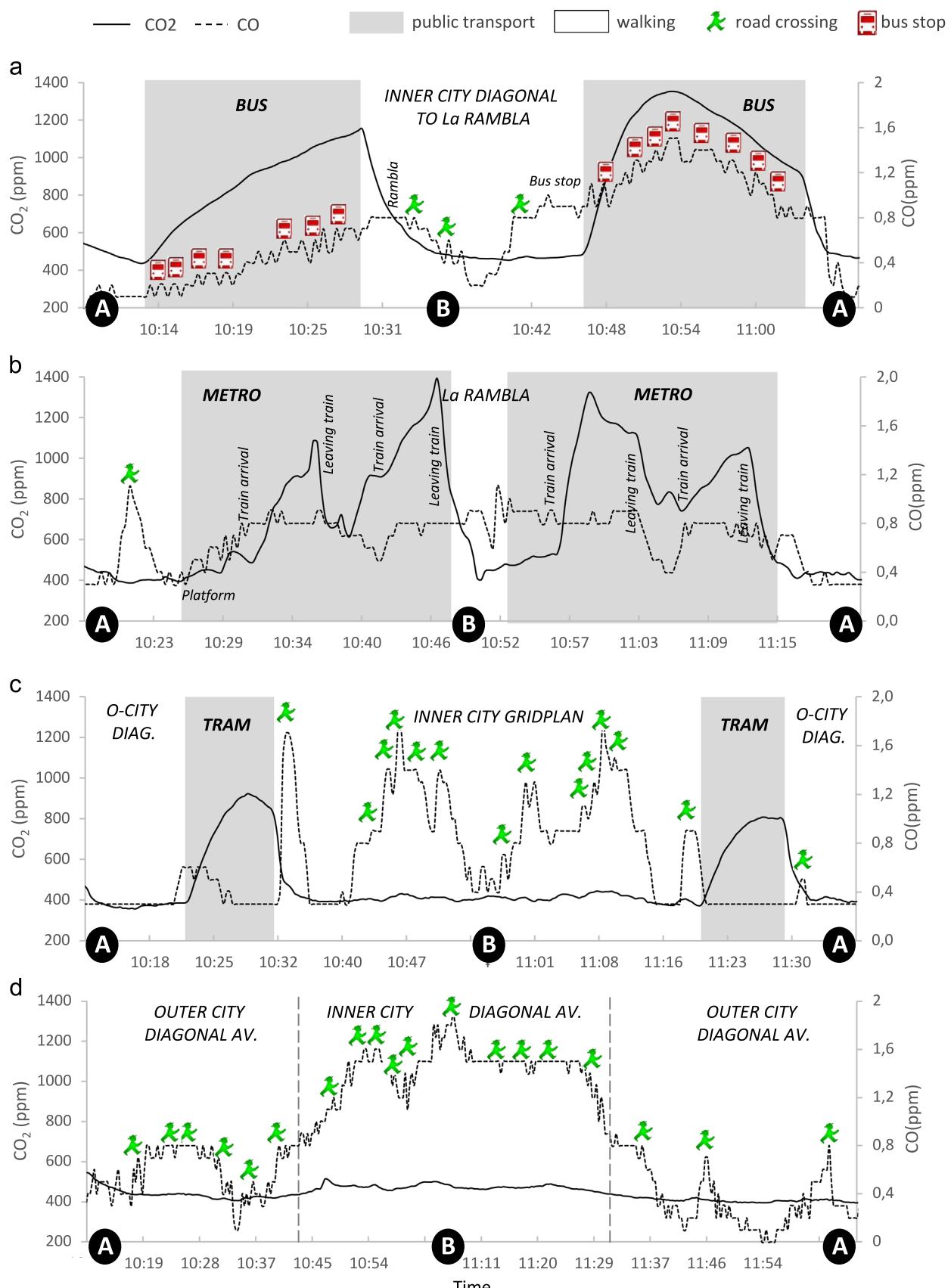


Fig. 5. GPS map of Black Carbon (BC) concentrations measured during the route done walking. See text for discussion.



**Fig. 6.** Distribution of CO and CO<sub>2</sub> concentrations during the route done by bus (a), metro (b), tram+walking (c) and walking (d). The time spent inside public transport is shown in grey. (A: start point at CSIC, B: final point at La Rambla).

**Table 3**Chemical composition of all PM<sub>2.5</sub> samples from commuting the same route walking, by metro or bus. Highest values of tracers are marked in bold. (dl: detection limit)

Date	Walking + works			Metro			Tram + walking			Bus			Average			
	6-14/10	15-23/10	24-31/10	6-14/10	13-21/11	24/11-2/12	24-31/10	5-12/11	13-21/11	15-23/10	5-12/11	24/11-2/12	Walking	Metro	Tram + walking	Bus
$\mu\text{g m}^{-3}$																
PM <sub>2.5</sub>	22.6	29.0	31.9	36.7	48.6	42.4	34.6	29.3	27.1	47.8	39.2	48.6	27.8	42.6	30.6	45.2
Al <sub>2</sub> O <sub>3</sub>	<b>1.0</b>	<b>1.6</b>	<b>0.9</b>	0.4	< dl	0.1	0.01	0.01	0.01	0.1	0.1	0.3	<b>1.2</b>	0.2	0.01	0.2
SiO <sub>2</sub>	<b>3.1</b>	<b>4.8</b>	<b>2.5</b>	1.1	< dl	0.4	0.03	0.03	0.03	0.2	0.3	0.8	<b>3.5</b>	0.5	0.03	0.4
CO <sub>3</sub> <sup>2-</sup>	2.0	3.2	2.7	3.0	0.7	1.3	1.40	0.51	1.03	2.4	1.9	1.1	2.7	1.7	0.98	1.8
Ca	1.3	2.2	1.8	2.0	0.5	0.9	0.94	0.34	0.68	1.6	1.2	0.8	1.8	1.1	0.65	1.2
Fe	0.8	1.1	1.5	<b>12.5</b>	<b>13.5</b>	<b>14.0</b>	0.86	0.66	0.71	2.3	2.3	2.0	1.1	<b>13.3</b>	0.74	2.2
K	0.4	0.5	0.5	1.5	< dl	0.5	1.18	1.19	0.40	1.3	0.6	< dl	0.5	0.7	0.92	0.6
Na	0.5	0.4	0.3	0.5	0.1	0.2	0.28	0.12	0.19	0.3	0.2	0.3	0.4	0.3	0.20	0.3
Mg	0.2	0.3	0.3	0.3	0.2	0.3	0.11	0.08	0.12	0.2	0.2	0.1	0.3	0.3	0.10	0.2
P	0.04	0.02	0.06	0.03	0.04	0.02	0.05	0.02	0.02	0.10	0.04	0.04	0.04	0.03	0.03	0.06
S	1.2	0.6	1.2	1.0	0.5	0.6	0.89	0.39	0.50	0.6	0.9	0.4	1.1	0.7	0.59	0.6
$\text{ng m}^{-3}$																
Li	< dl	0.80	0.83	< dl	< dl	< dl	< dl	< dl	< dl	< dl	< dl	< dl	0.80	< dl	< dl	< dl
Ti	<b>78.37</b>	<b>155.43</b>	<b>85.79</b>	16.20	20.43	44.68	3.97	11.51	27.56	21.95	49.02	32.99	<b>106.53</b>	27.10	14.35	34.65
V	11.64	8.86	6.27	4.91	4.20	4.87	4.32	3.26	2.95	3.89	2.49	16.78	8.92	4.66	3.51	7.72
Mn	13.59	22.06	19.48	<b>115.86</b>	<b>114.46</b>	<b>130.74</b>	15.97	14.39	12.34	34.82	21.28	18.01	18.38	<b>120.35</b>	14.23	23.71
Co	0.75	0.64	0.81	<b>1.67</b>	<b>1.28</b>	<b>1.57</b>	0.94	< dl	< dl	< dl	< dl	0.73	<b>1.51</b>	0.31	< dl	< dl
Cu	35.70	38.00	35.15	127.24	97.30	111.41	27.00	19.25	28.69	<b>177.25</b>	<b>166.51</b>	<b>168.18</b>	36.28	111.99	24.98	<b>170.65</b>
Zn	93.85	117.53	93.43	<b>186.27</b>	<b>173.13</b>	<b>180.23</b>	60.42	43.25	55.61	142.76	134.21	113.60	101.60	<b>179.88</b>	53.09	130.19
Ga	< dl	0.62	< dl	< dl	< dl	< dl	< dl	< dl	< dl	< dl	< dl	0.62	< dl	< dl	< dl	< dl
Ge	< dl	< dl	< dl	3.20	< dl	0.91	< dl	< dl	< dl	1.96	< dl	< dl	< dl	2.05	< dl	< dl
As	1.56	2.17	2.18	0.94	1.21	0.99	0.68	0.61	0.10	1.29	0.26	4.22	1.97	1.04	0.46	1.92
Rb	0.79	1.01	1.04	1.10	< dl	< dl	< dl	< dl	< dl	1.26	< dl	0.94	1.10	< dl	1.15	< dl
Sr	4.85	5.45	5.43	<b>15.93</b>	<b>12.27</b>	<b>14.57</b>	3.57	1.90	3.25	5.10	3.08	4.34	5.24	<b>14.26</b>	2.91	4.17
Y	2.54	2.18	2.86	3.12	0.45	3.83	2.03	2.25	3.31	2.97	3.33	< dl	2.53	2.47	2.53	3.15
Zr	8.13	6.44	8.49	20.95	5.67	< dl	< dl	0.04	< dl	40.35	13.32	43.43	7.69	13.31	< dl	32.37
Nb	0.06	0.37	< dl	< dl	< dl	< dl	< dl	< dl	< dl	< dl	< dl	0.11	< dl	< dl	< dl	< dl
Cd	0.19	0.22	0.23	0.13	0.02	0.31	< dl	< dl	0.29	< dl	0.21	< dl	0.22	0.15	0.29	< dl
Sn	5.10	5.92	6.49	6.06	5.07	5.14	4.81	3.36	3.49	6.59	4.30	6.11	5.84	5.42	3.89	5.67
Sb	1.90	2.14	1.90	3.02	1.89	2.51	0.52	0.38	1.08	<b>26.72</b>	<b>21.12</b>	<b>24.30</b>	1.98	2.47	0.66	<b>24.05</b>
Ba	26.97	21.66	20.60	<b>439.31</b>	<b>513.73</b>	<b>530.75</b>	22.55	19.71	17.15	56.73	53.76	69.08	23.08	<b>494.60</b>	19.80	59.86
La	0.49	0.52	0.61	0.26	0.24	0.63	< dl	< dl	0.48	0.18	1.10	0.20	0.54	0.38	0.48	0.49
Ce	1.08	1.18	1.41	0.67	0.54	1.49	< dl	0.08	1.08	0.46	2.47	0.46	1.22	0.90	0.58	1.13
Nd	0.26	0.34	0.27	0.04	< dl	0.39	< dl	< dl	< dl	0.13	0.18	< dl	0.29	0.22	< dl	0.16
Sm	0.69	0.69	0.83	1.02	< dl	1.13	< dl	< dl	0.94	0.90	1.14	< dl	0.73	1.09	0.94	1.02
Gd	0.19	0.15	0.17	0.22	1.06	0.31	0.06	0.08	0.25	0.15	0.23	< dl	0.17	0.52	0.13	0.19
Dy	0.47	0.42	0.59	0.66	0.04	0.74	0.41	0.49	0.71	0.60	0.62	< dl	0.49	0.47	0.54	0.61
Er	0.65	0.65	0.83	1.09	< dl	1.08	< dl	< dl	0.89	0.91	< dl	< dl	0.70	1.08	0.89	0.91
Pb	5.84	9.11	11.50	6.74	7.99	7.62	8.18	5.36	6.30	7.43	7.72	6.19	8.82	7.45	6.61	7.11
U	0.83	0.68	1.24	1.37	0.15	1.58	1.12	1.30	1.30	1.42	1.28	< dl	0.92	1.03	1.24	1.35

### 3.5. Carbon monoxide and dioxide

As would be expected, the distributions of carbon monoxide (CO) and carbon dioxide ( $\text{CO}_2$ ) show very different behaviours (Table 1). CO levels can clearly be related to traffic emissions, whereas  $\text{CO}_2$  levels are most influenced by the respiration of fellow travellers. Average CO levels were highest when walking in the inner Diagonal (1.1 ppm) and lowest while travelling inside the tram (0.4 ppm), with the rest registering a limited range between 0.7–0.9 ppm. In the case of  $\text{CO}_2$ , concentrations were highest in the bus (721 ppm), followed by metro (694 ppm) and tram (643 ppm), and much lower while walking (425–479 ppm). As would be expected, outdoor levels of  $\text{CO}_2$  levels remained stable at around 400 ppm (during the experiment seasonally adjusted atmospheric  $\text{CO}_2$  levels measured at the Mauna Loa Observatory in Hawaii exceeded 399 ppm in October 2014 for the first time).

Fig. 6 demonstrates the typical distribution patterns of these gases during the four commuting journeys. On entry into the (normally empty) bus both CO and  $\text{CO}_2$  begin a steady rise as the number of passengers increases and the bus interior becomes contaminated by traffic fumes from the Diagonal and probably from the diesel bus itself (Fig. 6a).  $\text{CO}_2$  levels immediately fall upon leaving the bus, whereas CO levels are maintained in the traffic-congested inner Diagonal area outdoors, falling only during the brief wait on the pedestrian La Rambla. On the return journey, when the bus is already busy and receiving more people as it travels along the traffic-congested inner Diagonal, both CO and  $\text{CO}_2$  rise to maximum levels ( $> 1 \text{ ppm}$  and  $> 1000 \text{ ppm}$  respectively). Beyond Francesc Macia the bus leaves the congested area and the number of passengers thins out, producing a steady decrease in  $\text{CO}_2$  until the end of the journey (Fig. 6a).

The relation between number of passengers and  $\text{CO}_2$  build-up is again seen when travelling by subway train, with a double peak separated by a trough reflecting the two segments of the rail journey separated by walking tunnel transfer (Fig. 6b). In this example  $\text{CO}_2$  over doubles in concentration during a short journey towards the city centre, reaching a peak of nearly 1400 ppm in the busy Diagonal station. On the return journey this same city centre peak declines in reverse pattern, although the final journey leg once again shows progressive passenger build-up of exhaled  $\text{CO}_2$  (in this case students travelling to the university stop at Palau Reial). In contrast CO levels remain stable at around 0.8 ppm inside trains, declining gently during the tunnel transfer. Travelling by tram similarly registers increasing  $\text{CO}_2$  with passenger number, rising from outdoor levels of around 400 ppm towards 1000 ppm, although with CO levels remaining very low except during the ingress of traffic fumes at some stops (Fig. 6c).

### 3.6. $\text{PM}_{2.5}$ chemical composition

Chemical analyses of filters collected during the experiment are shown in Table 3. The tram+walking mode samples show too much of a mixture and are not comparatively enriched in any chemical component. In contrast, notable differences can be identified between the pedestrian, bus and metro samples, expressed mainly in the amount of crustal ("geological") particles versus anthropogenic enrichments in metals. The unusually high amount of paving work activity during sampling along the inner Diagonal is reflected in relatively high concentrations of Al (and therefore calculated Si) and Ti, the latter element being three times more abundant in the Diagonal outdoor air than in the bus, or four times more than in the metro. For their part, the metro and bus samples each display their own distinctive mixtures of enriched metals. In the case of the metro this involves a cocktail of abundant iron accompanied by Mn, Co, Zn, Sr, Ba and Cu, whereas in the bus there are enhanced levels of Sb and, to a lesser extent,

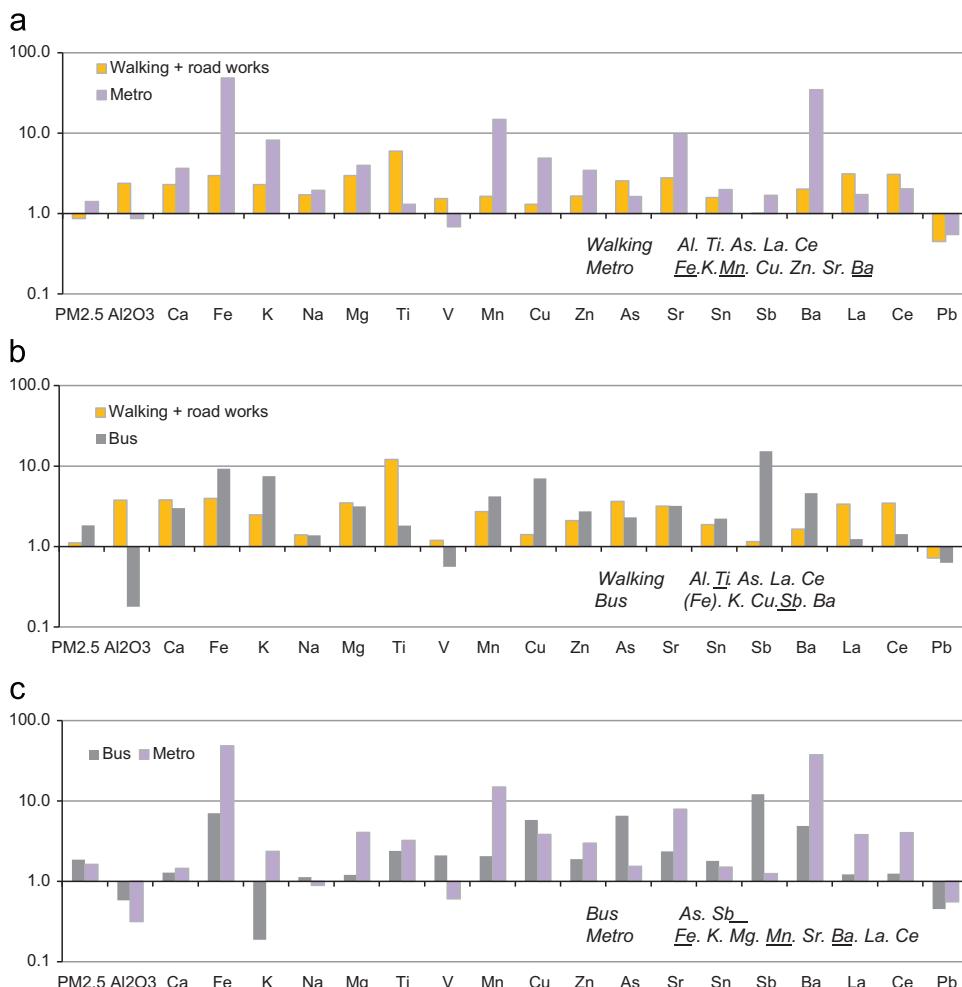
Cu and Ba, as typically shown in kerbside sites (Amato et al., 2014).

Fig. 7 graphically displays these various enrichments by comparing three pairs of samples, each collected simultaneously (7a: walking vs. metro; 7b: walking vs. bus; 7c: bus vs. metro). The analytical values for each pair are normalised to the average chemistry of  $\text{PM}_{2.5}$  in the urban background of Barcelona. In these normalised data the main metallic (Fe, Mn, Ba) enrichments for the metro environment, and that for Sb in the bus, can be seen to be over ten times greater than in the urban background environment outdoors (Fig. 7b,c). The likely sources of these metals will be discussed in the following section.

## 4. Discussion

Since studies reported earlier this century by pioneering publications such as Adams et al. (2001) and Kaur et al. (2006, 2007), data on urban air quality collected using mobile equipment have made it abundantly clear that individuals moving around a given city are constantly faced with ever-changing transient doses of atmospheric pollutants (e.g. Zuurbier et al., 2010; de Nazelle et al., 2012; Ragettli et al., 2013; Gu et al., 2015; Steinle et al., 2015). The effects of polluted urban air on human health are becoming increasingly apparent (e.g. Dominici et al., 2005; Ballester et al., 2008; Colais et al., 2009; HEI 2010; Knibbs et al. 2011; Pascal et al., 2013; Nyhan et al., 2014; Yang et al., 2015). Especially vulnerable are individuals already compromised by lung or heart dysfunction, such as asthmatics or citizens with heart rate variability (e.g. Liu et al., 2015 and references therein). We have reached the stage where this information needs to be passed to the individual commuter who has some choice in the matter. In this context, we will focus this discussion on two aspects: (1) the need for clear, easily interpretable information comparing the air pollution likely to be encountered on different commuting routes through a city; (2) the need to emphasise that "air quality" represents the sum of a range of physicochemical parameters (PM mass, size, number, chemistry; gas concentrations) the mixture of which will depend on the chosen transport route through the city and the area of the city where commuting occurs. In this context the results here discussed are specifically related to the area studied in the case of the walking route, but can be considered representative of equivalent transport modes used in other areas of the city and other urban environments. Note also that the values reported were measured during unexceptional atmospheric conditions for Barcelona, lacking any pollutant spikes that occur sporadically during the year due to anticyclonic stagnation or the arrival of dusty African air masses. In terms of average numbers of fine particles inhaled the following hierarchy of increasingly compromised air quality can be established: urban background < metro < tram < suburban main road walking < city centre walking < bus < city centre main road with paving works, with values for the metro being between two and three times less than those outdoors in the city centre (Table 1). As shown in Table 1 in the cases with higher N values, that is bus and walking in the city centre, mean values are higher than median, indicating that in these cases the distribution of N is not symmetric but skewed-right, this being related to the presence of few large values that increase the mean but do not affect the median. However, although such high peaks are influencing the mean values they should also be considered for exposure effects as transient pollutant peaks also affect commuter health.

Black carbon concentrations in the outdoor city environment serve, like N concentration, as a proxy for traffic pollution, particularly from diesel vehicles. Thus it is not surprising to find that the BC data record the same outdoor transient peaks as seen for N and  $\text{PM}_{2.5}$ , with values jumping up to 10 times higher when



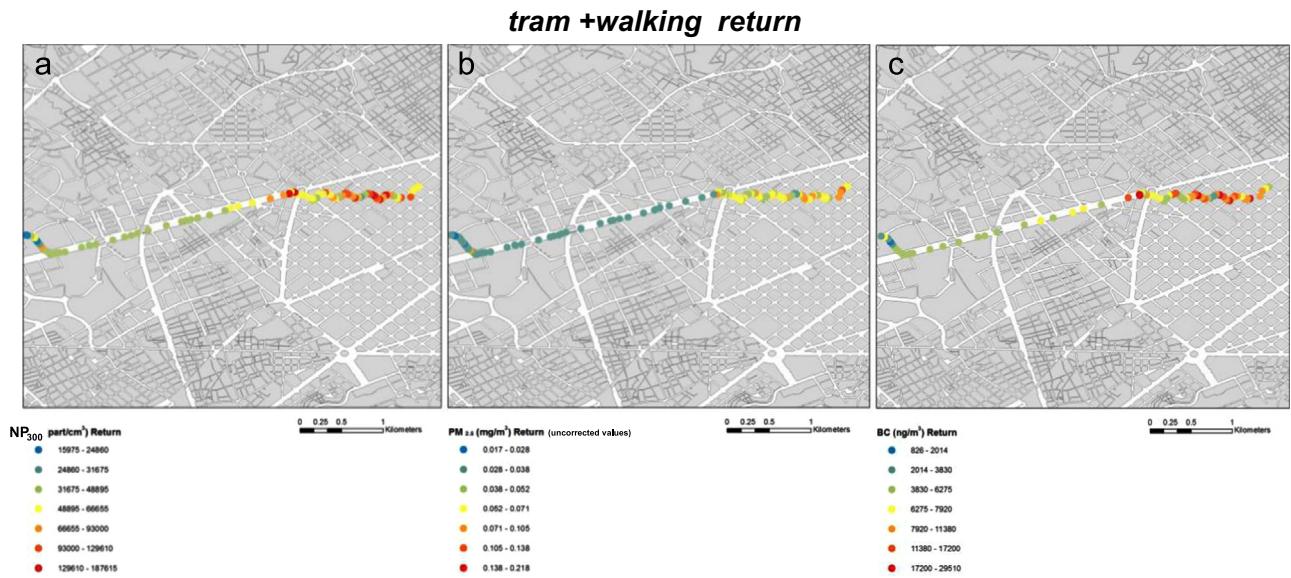
**Fig. 7.** PM<sub>2.5</sub> concentrations of selected major and trace elements analysed in samples taking coevally while walking or travelling by metro or bus. Values are normalised by the average concentrations measured in the Palau Reial Barcelona urban background monitoring site to show more clearly relative enrichments. Elements underlined are those more enriched for that transport mode.

crossing main roads such as Aribau or Muntaner in the city centre. In addition BC concentrations are also noticeably higher in the north side of the suburban (outer) Diagonal while walking (Fig. 5). As mentioned before this is attributed to the sea breeze influence during the day, bringing the traffic emissions to this side of the street (and not to the difference in traffic volume as the number of vehicles per hour at this point of the Diagonal is on average always higher at the end of the commuting trip: Fig. S2). As with the PM data, of the three public transport modes used the tram is the cleanest with respect to BC. The metro environment presents a special case in the measurement of BC, due to the high amount of ambient FePM which can interfere with the measuring equipment, therefore BC levels measured in this microenvironment are overestimated and should not be considered.

A third proxy for traffic contamination is provided by outdoor concentrations of CO, as illustrated in Fig. 6c and d. Levels of this gas in the city depend on wind direction and proximity to traffic: in the example shown walking along the north side of the Diagonal on the outward journey, with the wind blowing from the road, results in CO levels generally lower than those in the congested city centre, but higher than on the return (south side) suburban journey where the wind direction diluted traffic contamination (Fig. 6d).

Figs. 2–6 offer examples of how comparative air quality data can be presented graphically in a clear and straightforward manner. In terms of particle number concentration (Figs. 2–3),

travelling by diesel bus (with filter traps) or walking through Barcelona city centre are both obviously less attractive options than moving around underground by metro train. A similar conclusion, focussed specifically on health, has recently been reached by a study in Taiwan by Liu et al. (2015) who observed the least effects on heart rate variability in those commuters choosing the Taipei subway as their preferred form of city transport. When considering PM mass, however, differences between metro and bus travel are less evident (Fig. 4a,b), presumably due in part to the heavier, more ferruginous nature of ambient subway particles (Moreno et al., 2015 and references therein). Interestingly, the data on fine particle mass concentrations underground also reveals that not all train lines have the same level of contamination, with the segment travelled along Line 3 showing consistently higher PM concentrations than Line 5 (Fig. 4b), even though both lines have similar ventilation. Outdoors, dramatically fluctuating mass concentrations mirror those recorded by particle numbers and similarly reflect traffic patterns, although with the additional contribution of inhalable PM released by road and paving works. The air quality measured in the entire two months campaign, at least with regard to fine particle mass, was worst within the inner city (Diagonal and L'Eixample gridplan). Along the inner Diagonal a combination of dense traffic movement punctuated by frequent traffic light crossings, bus and taxi lanes, medium-rise architecture, and pervasive paving works during the restructuring of the entire avenue, all contributed to making this an especially



**Fig. 8.** GPS map of  $N$  (a),  $PM_{2.5}$  (b) and  $BC$  (c) concentrations measured during the route done by tram+walking. See text for discussion.

**Table 4**  
Air quality hierarchy based on nanoparticle concentrations

**Level 1 ( $< 1 \times 10^4$ part. cm $^{-3}$ )**

Clean outdoor and indoor environments free from traffic and cooking influences<sup>a</sup>

**Level 2 ( $1\text{--}2.5 \times 10^4$ part. cm $^{-3}$ )**

Low indoor<sup>b</sup> and outdoor<sup>a</sup> urban pollution, rural commuting by over ground trains (e.g. schools<sup>c</sup>; city suburban walking<sup>d,e</sup>; subway commuting in Barcelona<sup>f</sup>, London<sup>g</sup>, Stockholm<sup>h</sup>, Hong Kong<sup>i</sup>); bar after enactment of the anti-smoking law<sup>j</sup>

**Level 3 ( $2.5\text{--}5 \times 10^4$ part. cm $^{-3}$ )**

Moderate urban pollution (e.g. city walking<sup>f,k,l,m</sup>, tram<sup>f</sup>, bus<sup>i,k,n,o,p,d</sup> and metro commuting<sup>p,r</sup>; light indoor cooking<sup>s</sup>)

**Level 4 ( $5\text{--}10 \times 10^4$ part. cm $^{-3}$ )**

High urban pollution (e.g. walking<sup>e,f,t,u,v</sup> and cycling<sup>l</sup> in city centre, bus<sup>f,l,v,w</sup> and car<sup>k</sup> commuting; cooking in bars and restaurants<sup>x</sup>)

**Level 5 ( $> 1.0 \times 10^6$ part. cm $^{-3}$ )**

Extreme combustion peaks (e.g. proximal traffic plumes; indoor cooking<sup>a</sup>; use of butane heaters), bus<sup>t</sup> and taxi<sup>l</sup> commuting

<sup>a</sup> Buonanno et al. (2014b).

<sup>b</sup> Buonanno et al. (2011).

<sup>c</sup> Rivas et al. (2014).

<sup>d</sup> Ragettli et al. (2013).

<sup>e</sup> McCreanor et al. (2005).

<sup>f</sup> This study.

<sup>g</sup> Seaton et al. (2005).

<sup>h</sup> Midander et al. (2012).

<sup>i</sup> Yang et al. (2015).

<sup>j</sup> Pey et al. (2013).

<sup>k</sup> Mackay (2004).

<sup>l</sup> Kaur et al. (2006).

<sup>m</sup> Quirós et al. (2013).

<sup>n</sup> Levy et al. (2002).

<sup>o</sup> Zuurbier et al. (2010).

<sup>p</sup> Suárez et al. (2014).

<sup>q</sup> Knibbs et al. (2011).

<sup>r</sup> Aarnio et al. (2005).

<sup>s</sup> Gu et al. (2015).

<sup>t</sup> Kaur et al. (2005).

<sup>u</sup> Kaur et al. (2007).

<sup>v</sup> de Nazelle et al. (2012).

<sup>w</sup> Knibbs and de Dear (2010).

<sup>x</sup> Buonanno et al. (2010). Also included own (unpublished) data from Barcelona and Sitges (NE Spain).

polluted linear route through the city. At the time of writing the major works along this route have been completed and air quality has presumably improved.

Levels of CO record the presence of more heavily road trafficked zones ( $> 1$  ppm) as opposed to more suburban, pedestrian and underground metro environments, and thus reinforce the  $N$  data in emphasising the pervasive presence of traffic fumes in the city. Such data demonstrate how, while the pollution from traffic emissions is clear enough in the inner Diagonal, it is even higher in the gridplan L'Eixample, despite this being considered a highly favoured area to live in Barcelona (Figs. 3d and 6d). Traffic emissions do not much influence the air quality when travelling inside the tram (Fig. 8), where concentrations of  $N$ ,  $PM_{2.5}$  or  $BC$  are consistently low. However walking in the gridplan area immediately raises exposure to all these parameters, this being especially pronounced in the streets running NE-SW such as Casanova, Muntaner or Aribau, with a hill-climbing topography favouring more intense traffic emissions (Fig. 8). Commuting in one of the more heavily congested streets in this central zone, especially with traffic climbing from sea to mountain, is a lifestyle choice with no consideration for the possible health effects of poor air quality. In contrast, outdoor levels of  $CO_2$  are unaffected by city centre congestion, although they can triple inside a crowded bus or metro train, reaching levels over 1200 ppm (Fig. 6).

The values here reported while commuting by public transport or walking are generally lower than those reported by other works while commuting by private car (Karanasiou et al. 2014, and references therein). Higher exposure to PM (Boogaard et al., 2009), BC (Adams et al., 2001; de Nazelle et al., 2012) and UFP (Ragettli et al., 2013) concentrations are reported when travelling by car compared to other transport modes, although this is highly dependant on the traffic intensity, the ventilation inside the vehicle or the fuel type (Rank et al., 2001; Diapouli et al., 2008; Cattaneo et al., 2009; Geiss et al., 2010; Jalava et al., 2012). Thus reported air pollutant concentrations inside cars vary between 22 and 85  $\mu gPM_{2.5} m^{-3}$ , 6–30  $\mu gBC m^{-3}$ , whereas  $N$  can exceed  $3 \times 10^4$  cm $^{-3}$  (Alm et al., 1999; Dennekamp et al., 2002; McNabola et al., 2009; Boogaard et al., 2009; Cattaneo et al., 2009; Geiss et al., 2010; Zuurbier et al., 2010; Dons et al., 2011; Kingham et al.,

2013).

A user-friendly method of comparing air quality is to define hierarchies of increasing pollution during commuting exposure, such as the one we introduce in Table 4. Here we combine published data on particle number concentration with our own database from Barcelona city and the much less polluted coastal town of Sitges, 40 km to the SW. In this Table air quality is classified according to a simple scale that deteriorates across an order of magnitude from **Level 1 (Clean:  $< 1.0 \times 10^4 \text{ part. cm}^{-3}$ )** to **Level 5 (Extreme:  $> 1.0 \times 10^5 \text{ part. cm}^{-3}$ )**, with thresholds being selected to mark the difference broadly between low, moderate and high levels of traffic-related pollution typically found in the urban environment. Level 1 (Clean) conditions characterise outdoor environments away from traffic and are unlikely to be encountered in central Barcelona, although they can be found in traffic-free parts of Sitges (seafront, central pedestrian zones, well-ventilated shops) as well as in overground trains passing through the more rural part of the journey from Sitges to Barcelona. **Level 2 (Low Urban Pollution:  $1.0 \times 10^4\text{--}2.5 \times 10^4 \text{ part. cm}^{-3}$ )** conditions are common in quieter urban streets with relatively low background levels, and include average  $N$  measured in the Barcelona metro during our study. **Level 3 (Moderate Urban Pollution:  $2.5 \times 10^4\text{--}5.0 \times 10^4 \text{ part. cm}^{-3}$ )** we identify as typical of many urban streets with high to moderate traffic flow, and includes indoor tram air, some bus and subway commuter routes, as well as conditions commonly found in bars and restaurants. **Level 4 (High Urban Pollution:  $5.0 \times 10^4\text{--}1.0 \times 10^5 \text{ part. cm}^{-3}$ )** represents the concentrations of ultrafine particles commonly encountered outdoors in heavily trafficked city centres such, inside many buses, diesel trains in tunnels, as well as indoor areas significantly contaminated by cooking emissions. Finally **Level 5** conditions are found in the more extreme traffic hotspots, whether walking or driving, in poorly ventilated road tunnel air, and associated with uncontrolled cooking emissions or hydrocarbon combustion plumes such as those close to diesel exhaust pipes or butane gas heaters (see references in Table 4).

An unusual aspect of this commuting study in Barcelona is that it includes chemical characterisation of the air breathed on each of the four routes. As far as we are aware, this is the first time personal exposure to aerosol chemistry has been tracked through a city using different commuting routes. Given the fact that to obtain just one “pedestrian-only” sample for chemical analysis it was necessary to walk over 20 km, this is perhaps not surprising. Despite the reduced number of samples obtained, our pilot study demonstrates interesting variations in ambient air depending on the route taken. Commuters choosing to walk all the way to the city centre are those most likely to inhale not only fresh traffic fumes but also more “crustal” particles, i.e. those comprising natural rock-forming minerals such as quartz, feldspars and clays. Such materials ultimately will have been eroded from rocks and soils and resuspended by wind and passing traffic, and be enriched in Al and Si and their accompanying characteristic “geologically-derived” trace elements such as Ti (Moreno et al., 2006). In contrast, metro system air has an entirely different, ferruginous, character. The ambient FePM that dominates such air is typically accompanied by a highly specific mix of trace metals such as Mn derived from the steel, and Ba sourcing from brakes (Querol et al., 2012; Martins et al., in press; Moreno et al., 2015). Ambient inhalable PM in buses can also display its own distinctive metallic trace element geochemistry. In the case of Barcelona diesel bus route 33 involves a clear enrichment in Sb and Cu. The source for these elements is probably also from the braking system as shown at kerbside sites by Amato et al. 2014, although this requires further investigation.

To illustrate graphically these distinctive chemical differences we introduce a MnTi(Sbx10) ternary plot on which the differing

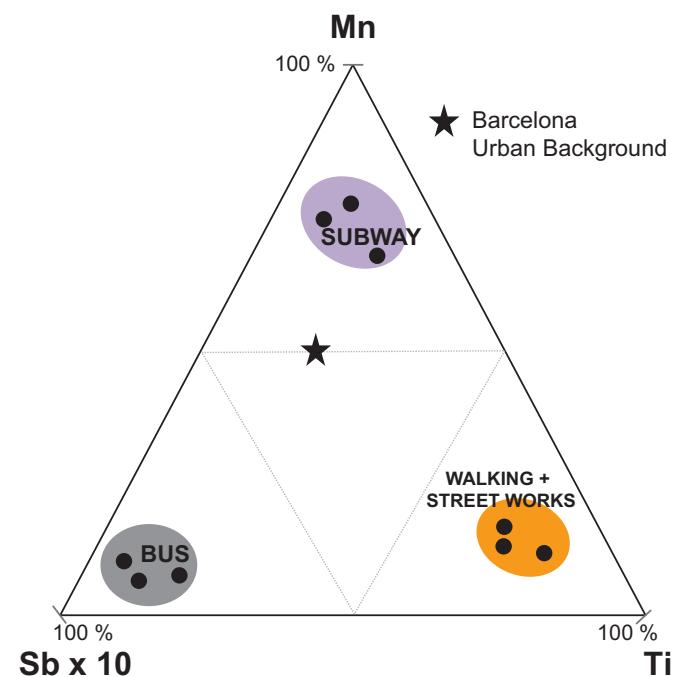


Fig. 9. Chemical differences between commuting transport modes, using key pollutant tracers.

chemistries of ambient PM breathed on the three commuting routes can be clearly separated (Fig. 9). In addition to physical variations in ambient PM, significant chemical differences can often also be identified, as for example demonstrated in previous publications on CuZnPb aerosols in Mexico City, La-contamination from refineries, and the cocktail of metals released during recreational firework displays (Moreno et al., 2007, 2008, 2010). To this list we can now begin to add variations in inhalable aerosol chemistry encountered in different typical urban commuter transport systems. According to our study, for example, the urban traveller may inhale perhaps ten times more Ti or Mn or Sb during the daily commute, depending on the route chosen to move through the city.

## 5. Conclusions

- (1) Urban air includes many microenvironments to which the citizen is exposed at different times of the day, with the air quality of most of them being very different from that measured at urban background monitoring sites. Our study compares tram, metro, bus and walking routes through Barcelona and reinforces understanding of how real-time air pollutant concentrations regularly inhaled by urban commuters vary greatly depending on how they choose to travel. The “average air quality” encountered during a city journey involves several different physicochemical parameters, not all of which will rise and fall in accordance with each other as they are linked to different sources and processes.
- (2) We define urban pollution ultrafine particle levels as rising through a scale of five levels: L1 (Clean), L2 (Low), L3 (Moderate), L4 (High) to L5 (Extreme). The average particle number concentrations measured are lowest in the commute using subway trains ( $N < 2.5 \times 10^4 \text{ part. cm}^{-3}$ =Level 2) and highest in those using diesel bus or walking in the city centre trafficked streets ( $N > 5.0 \times 10^4 \text{ part. cm}^{-3}$ =Level 4). Pedestrians at busy city centre traffic crossings are not uncommonly exposed to

- ultrafine particle transient peaks reaching Level 5 on this scale ( $> 1.0 \times 10^5 \text{ part. cm}^{-3}$ ).
- (3) Alveolar deposited surface area concentrations are lowest in the tram and metro, and highest when travelling by bus. Furthermore, the subway displays a particle size mode larger (90 nm) than in outdoor commuting environments ( $< 70 \text{ nm}$ ).
  - (4) With regard to ambient concentrations of fine particulate matter (PM<sub>2.5</sub>) however, commuting using the tram appears to be consistently the cleanest form of city public transport when compared to both bus and subway.
  - (5) Outdoor CO concentrations (as with Black Carbon) offer a good proxy for traffic contamination, whereas CO<sub>2</sub> concentrations are a good indicator of the degree of indoor passenger crowding on public transport.
  - (6) The chemistry of inhaled PM<sub>2.5</sub> varies markedly depending on the mode of city travel adopted. In addition to their direct exposure to traffic emissions, urban roadside pedestrians can inhale more siliceous "crustal" dust derived from rocks and soils, whereas subway passengers inhale a more obviously anthropogenic particle mix enhanced in Fe, Mn, Co, Zn, Sr and Ba. As described for kerbside sites bus air registered unusually high levels of Sb and Cu, typical tracers of brake wear.
  - (7) The kind of data reported in this paper, obtained using mobile equipment pairs simultaneously monitoring and comparing contrasting commuting journeys on a given day, aims at providing the urban traveller interested in air quality with better informed choices as to how best to minimise air pollution negative health effects when moving around the city.

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## Appendix A. Supplementary material

Supplementary data associated with this article can be found in the online version at <http://dx.doi.org/10.1016/j.envres.2015.07.022>.

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