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Fine particle concentrations in buses and taxis in Florence, Italy

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ABSTRACT

On October 2004, a sampling survey was carried out in Florence to estimate urban fine particle exposure concentrations inside commuting vehicles during workdays characterized by heavy traffic. Portable samplers were positioned inside four regularly scheduled diesel-powered buses and four taxis during eight weekdays. Each sampler consisted of a 2.5 μm size pre-separator cyclone, a direct-reading data logging photometer (pDR-1200), and a 4 L min^{-1} filter sampler for the determination of $PM_{2.5}$ mass concentration. Based on reflectance analysis measurements, a $PM_{2.5}$ Black Smoke Index was determined for each filter, and the elemental composition of the $PM_{2.5}$ was analyzed by Particle Induced X-ray Emission (PIXE). $PM_{2.5}$ mass concentrations inside the vehicles correlated well with the urban ambient air $PM_{2.5}$ concentrations measured at the fixed-site monitoring stations. The $PM_{2.5}$ excess above the urban ambient level was on average 32 μg m^{-3} (range: 22–52 μg m^{-3}) and 20 μg m^{-3} (range: 11–29 μg m^{-3}) in buses and taxis, respectively. The $PM_{2.5}$-bound sulfur concentration was also higher in the buses than in the taxis. Based on daily Time-Microenvironment-Activity-Diary (TMAD) data, the Florentines spend on average 9.7% of their day in traffic, and the corresponding average exposure is approximately 12% of their daily $PM_{2.5}$ personal exposure. The obtained data could be used to plan interventions to minimize the $PM_{2.5}$ citizen exposures in commuting.

1. Introduction

Epidemiological studies (US-EPA, 2004; WHO, 2005; Pope et al., 2002, 2004) have associated adverse and severe health outcomes with exposure to ambient air Particulate Matter less than 2.5 μm in aerodynamic diameter ($PM_{2.5}$), especially among susceptible sub-populations, e.g., infants, the elderly, and subjects with cardiopulmonary diseases.

In urban areas, a large fraction of the $PM_{2.5}$ pollution originates from local mobile sources: mainly diesel-powered vehicles and twin stroke engine powered motorcycles (Kinney et al., 2000), and stationary combustion processes (heating and industrial). $PM_{2.5}$ from local and micro-environmental sources is superimposed on a varying regional background $PM_{2.5}$, which consists of primary particles, originating from combustion emissions and soil dust, and secondary aerosols (ammonium sulfate and nitrate, and organics).

Florence (102.3 km^2, and 367 000 inhabitants) is located in Central Italy. It is characterized by cold winters and hot

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summers due to its location, inland from the basin of the River Arno. The city center is ancient and has very narrow street canyons. The primary domestic heating fuel is natural gas. The urban traffic-related air pollution of Florence has been estimated to account for about 40% of ambient PM_{10} pollution (Lucarelli et al., 2004). Each day, more than 139 000 trips within the city, about 30 000 trips from the city to the industrial fringe municipalities, and about 107 000 trips to the city from neighboring residential communities are made for work, shopping, and leisure (Viviani et al., 2005). Moreover, due to its exceptional historical and artistic attractions, many tourists travel to and from the city each day. Therefore, the daily number of commuters is large with subsequent traffic congestion.

The Florentine share of private cars in commuting is 38.6%, followed by motorcycles (21%) and urban public buses (14.6%). Most of the ultra-fine particles are emitted from on-road older diesels and motorcycles with no particle filters or catalysts; meanwhile, the contribution from the most recent gasoline-powered vehicles is small. (http://www.comune.fi.it/servizi_pubblici/ambiente/convegnoinquinamento/04RelazioneGrechi-Galanti.htm).

The study's aim was to characterize the $PM_{2.5}$ mass levels, the $PM_{2.5}$-bound element concentrations and the Black Smoke (BS) index values inside buses and cars (taxis) in typical workday traffic conditions. We report the modeling method and the estimated $PM_{2.5}$ exposures for Florentine non-smoking citizens commuting on urban routes to improve understanding of the impact of the time spent in traffic.

2. Materials and methods

2.1. Bus survey

The local public transport joint-stock company is a consortium of several municipalities of the Florentine metropolitan area. Its service covers a total surface area of 538 km^2. In 2004, the company transported 82 million passengers and its bus operating fleet consisted of 443 buses with an average age of 8 years: 66% fuelled with Ultra-Low Sulfur Diesel (ULSD), 14% with Compressed Natural Gas (CNG), 14% with *Gecam*™, and 6% with electricity. In the urban area, the buses travel on 33 fixed major routes: 16 of them radially linking the historic center to the surrounding municipalities, two circular routes on the ring boulevards, four routes within the historic center (all electrical), four tangential by-passing the center, and seven diametrical connecting densely-populated neighborhoods across the historic center.

Orange diesel buses ($n=4$) traveling on in-use urban routes were chosen for the sampling campaign (Table 1) by age, fuel, manufacturer, and street coverage by traffic density as representatives of the operating bus fleet. The sampling was carried out on October 4, 6, 8, and 11, 2004. A Portable Exposure Monitor (PEM) was placed in each bus approximately 1 m above the bus floor, within 0.5 m of the driver, and at the height of the driver's breathing zone. There was no dividing wall between the driver and the passengers. During the daily sampling time (e.g., from 6 a.m. to 9 p.m.), each bus was engaged in its regular workday service, route and timetable exactly as usual. The selected buses had vertical exhaust pipes located on the roof at the rear of the bus and the air-conditioning systems were off. The drivers were told to drive and ventilate the bus as usual.

Smoking on public transport system has been prohibited by the Italian Law no. 584 since November 11, 1975.

2.2. Taxi survey

The sampling was carried out in taxis ($n=4$) on October 13, 15, 18, and 20, 2004 from approximately 8:30 a.m. to 8:00 p.m. The taxis were out of traffic during the lunch breaks and while waiting for customers and cruised mostly within the city. As with the buses, the drivers were told to drive and ventilate the car as usual. The taxi drivers were non-smokers. During the sampling, the drivers completed a specific TMAD, recording the time when they opened the windows, stopped the car, used the air conditioning, or drove out of the urban zone. The PEM was always located near the hand brake close to the driver. Table 1 presents the taxi characteristics. During the sampling, the drivers did not start, wait, stop or park inside closed garages.

2.3. The 24 h-TMADs and personal exposure to $PM_{2.5}$ in commuting

Workday 24 h-TMADs from a non-random sample of healthy non-smoking volunteers ($n=38$; age 21–67 years old), out of 654 interviewed people (by phone) living in the inner-city quarters of Florence, were drawn for the study on personal exposure to $PM_{2.5}$ during commuting. They were first asked about their time spent in traffic and their ability to participate in the personal exposure survey performed in October–December 2004. Each volunteer filled in the 24 h-TMAD (15 min resolution) while his/her 24 h-personal $PM_{2.5}$ exposure was measured with a PEM. The microenvironment categories were in transit (walk, bike, motorbike, car/taxi, bus, and train), waiting (in and out), and other (home: in and out, work/school: in and out, and others: in and out). They also registered passive smoking exposure (defined by the presence of at least one smoker within 5 m). The 24 h-TMADs and on-board vehicle mass concentrations were used to estimate the average daily partial personal $PM_{2.5}$ exposure in commuting for urban trips by car or bus.

The E_i is calculated from the sum of daily average exposures for the m total in-commuting microenvironments encountered by one person i during the average time for exposure (24 h).

$$E_i = \sum_{j=1}^{m} Eij = \sum_{j=1}^{m} \left(1/T \sum_{k=1}^{n} Cijk \cdot tijk \right)$$

Where for one person i: E_i = contribution of in-commuting $PM_{2.5}$ to daily average exposure ($\mu g\, m^{-3}$). Eij = contribution of each commuting microenvironment j to the daily average ($\mu g\, m^{-3}$). T = average time for exposure (24 h). $Cijk$ = $PM_{2.5}$ exposure concentration in the in-commuting

Table 1
Characteristics of vehicles and routes

Vehicle	Vehicle model	Fuel type	Emission standard	Registration year	Urban route type
Bus	BredaMenarinibus BMB 240	ULSD[a]	Euro 3	2001	70 passengers/trip, line length = 23.5 km with 98 stops
	BredaMenarinibus 10 M city	ULSD[a]	Euro 2	2001	Diametric[c]; 52 passengers/trip; line length = 17.3 km with 53 stops
	BredaMenarinibus 12 metri	ULSD[a]	Euro 0	1983	Diametric[c]; 95 passengers/trip; line length = 24 km with 82 stops
	BredaMenarinibus 12 metri	ULSD[a]	Euro 3	2002	Diametric[c]; 66 passengers/trip; line length = 22 km with 75 stops
Taxi	Fiat Marea SW	Diesel	Euro 3	2002	Random route
	Fiat Multipla	Bi-fuel: CNG[b] & unleaded gasoline	Euro 3	2000	Random route
	Fiat Multipla	CNG[b]	Euro 3	2000	Random route
	Fiat Multipla	CNG[b]	Euro 3	2000	Random route

[a] ULSD = Ultra-Low Sulfur Diesel (sulfur less than 10 ppm).
[b] CNG = Compressed Natural Gas.
[c] Crossing the city center and connecting suburban areas.

microenvironment *j*. *tijk* = time spent inside in-commuting microenvironment *j* for the *k*-th time.

2.4. Sampling and analytical methods

The PEMs ($n = 4$), previously used in the European ULTRA Study (Janssen et al., 2000), were used for the vehicle and personal $PM_{2.5}$ sampling. Each PEM consists of a 4 L min^{-1} personal sampling battery-operated pump (BGI 400, BGI Inc, Waltham, MA, USA), a $PM_{2.5}$ GK2.05 (KTL) cyclone (BGI Inc., Waltham, MA; device conforms to US-EPA $PM_{2.5}$ Standard when operated at 4 L min^{-1}), a standard 37 mm plastic filter cassette with a 2 μm pore size pre-conditioned and pre-weighed ringed Teflon (PTFE)-membrane filter (Teflo® R2PJ037, Pall Gelman Sciences, MI, USA), and a real-time aerosol monitor/data logger Personal DataRAM™ (pDR, model 1200; MIE Inc, MA, US). The pDR is an integrating nephelometer that continuously measures the amount of light ($\lambda = 880$ nm) scattered by particles drawn through a sensing zone (accuracy: ±5%, resolution 0.001 mg m^{-3}). It is capable of measuring short-term exposure levels and concentration profiles during the sampling periods. One PEM did not have the pDR instrumentation. Before each survey day, the pDR was set to zero in a smoke-free room using HEPA-filters provided by the manufacturer. The accuracy of the airflow was checked before and after each sampling with a bubble flow meter as a primary standard (Buck M-30 by A.P. Buck Inc. Orlando, FL, USA). The PEM preparation, calibration, and directions for use, as well as the gravimetric filter analysis, were carried out according to the standard operation procedures of the ULTRA study (Janssen et al.; 2000). Tiny Tag Ultra data loggers (Gemini Loggers Ltd, UK) were attached to the PEM cases to measure site-specific Temperature (T) and Relative Humidity (RH%) on-board vehicles. These data were recorded continuously and downloaded after the sampling.

The filters were equilibrated for at least 24-h before weighing. Filters were loaded and unloaded onto the filter holder in a weighing room, sheltered from vibrations and drafts, with controlled T and RH% ($T = 20.7 \pm 1.2$ °C; RH = 50 ± 10%) to minimize the particle volatilization and aerosol water bias. Air flow rates were calibrated, checked and recorded at the beginning and end of each sampling period to eliminate those samples with a flow rate changing more than ±5%. A One-Point-Ionizer (OPI, HAUG GmbH & Co. KG Leinfelden-Echterdingen, Germany) was used to minimize weighing errors induced by electrostatic charge. Each filter was weighed twice using a 1 μg reading micro balance (Mettler MX 5, Mettler-Toledo Ltd., Greifensee, Switzerland), and this was repeated if the difference between the two measurements exceeded 1 μg. Before the weighing sessions, the microbalance was calibrated with certified stainless steel weights (100 mg) to further validate the internal calibration. After weighing, the samples were stored in sealed Petri dishes in a freezer for further analyses.

$PM_{2.5}$ reflectance was measured, using a Smoker Stain Reflectometer (M43D, Diffusion Systems Ltd, London, UK), in the center and in the four quadrants of each sampled filter in a dark room; the obtained mean value was transformed into the $PM_{2.5}$ specific absorption coefficient (BS index) using the ISO 9835 formula (ISO, 1993).

The concentration of elements was measured by the PIXE analysis, using the 3 MV Tandetron accelerator at the LABEC laboratory of INFN, with the external beam set-up extensively described elsewhere (Chiari et al., 2005). PIXE spectra were fitted using the GUPIX software package, and elemental concentrations were obtained via a calibration curve from a set of thin standards of known areal density. Mean elemental $PM_{2.5}$ concentrations in the ambient air were then calculated using the sample deposition area on the filter, air flow rate and sampling duration. The associated uncertainty was determined by a sum of independent uncertainties on standard certified concentrations (5%), peak areas (from 2 to 20% or higher when concentrations approach the detection limit), which include X-rays counting statistics, background subtraction and peak overlaps, aerosol deposition area and air flow rate (few percent). The following elements were traced: Na, Mg, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, As, Se, Br, Sr, Zr, Mo, Ba and Pb. The accuracy of the PIXE analysis was confirmed with a National Institute of Standards and Technology standard. Blank filters

were analyzed to detect background elements to be subtracted. The elemental concentrations under the Minimum Detection Limit (MDL) were replaced with their MDL/2 before the arithmetic mean calculation.

2.5. Meteorological factors and ambient air $PM_{2.5}$

Hourly averages of ambient *T*, RH%, pressure, precipitation, and wind speed and direction during the sampling days were obtained from the Tuscan Meteorological Centre Osservatorio Ximeniano, located in the center of Florence.

The impacts of wind speed and rain (mm) on the $PM_{2.5}$ mass and composition were analyzed. In Florence, the 24-h $PM_{2.5}$ measurements, according to the European Directives, were performed from 2000. The ambient urban $PM_{2.5}$ was sampled alternatively with PM_{10}, each for 15 days, in all sites of the public air quality-monitoring network. The fixed-site monitoring stations are equipped with Beta Gauge Monitors, (model MP101 M or MPSI 100, Environment s.a., France), managed by the Tuscan Environmental Protection Agency (ARPAT).

We used data from four air quality-monitoring sites. The first two sites are classified as "urban background" and the other two as "traffic-oriented" sites (European Commission decision 2001/752/EC). For each sampling day, the urban median value of the measured or estimated (from PM_{10}) 24-h $PM_{2.5}$ concentrations was used as an estimate of the general population exposure to $PM_{2.5}$.

2.6. Statistical analysis

Arithmetic Means (Ms) and Standard Deviations (SDs) were calculated for gravimetric mass and pDR $PM_{2.5}$ concentrations, BS index and elemental concentration data. The Wilk–Shapiro normality test and the *P*-probability plot correlation test (Filliben test) were used to determine the normality of the data. The Median and the Kruskal–Wallis tests were used to compare the $PM_{2.5}$ concentrations measured in the two groups of vehicles. The relationship between meteorological factors and the measured concentrations were characterized by Spearman's (ρ_s) and Pearson's (r) correlation coefficients, and the adjusted determination coefficient (R^2-adj).

Elemental concentrations were used to roughly estimate the contribution of different sources to $PM_{2.5}$ by multiple linear regression, with the $PM_{2.5}$ mass concentrations as the dependent variables and BS index (marker of local traffic component), S concentration (marker of secondary sulfate and local primary diesel PM) and Si concentration (marker of the soil component) as independent variables. The mass of the soil component was also calculated considering elements of crustal origin as oxides (Miranda et al., 1994).

The statistics were performed using Intercooler STATA software (version 8.0, STATA Corporation, College Station, Texas).

3. Results

The mean sampling time for the buses was 15.1 h (range: 14.0–15.5 h) with each single commuting trip time ranging from 38 to 54 min. The taxi mean sampling period was 11.2 h (range 9.7–12.2 h).

Table 2 displays the some statistics obtained for gravimetric, real-time analysis (pDR) and BS index data, by type of vehicle. $PM_{2.5}$ mass concentrations inside the buses ranged between 33 and 77 $\mu g\,m^{-3}$, with a M(SD) equal to 56(15) $\mu g\,m^{-3}$. In the taxis, the $PM_{2.5}$ mass concentrations ranged between 20 and 70 $\mu g\,m^{-3}$, with a M(SD) of 39(15) $\mu g\,m^{-3}$. However, also the urban background $PM_{2.5}$ concentration differed between the dates of the buses 29(12) and of the taxis 19(12) $\mu g\,m^{-3}$ measurements.

Low wind speeds (average below 2 $m\,s^{-1}$) during the bus survey days increased the ambient $PM_{2.5}$ concentrations, while the higher than 5 $m\,s^{-1}$ speed for two taxi survey days decreased the ambient $PM_{2.5}$ during the taxi sampling time. Temperature in the buses and taxis ranged from 21 to 27 °C, and RH ranged from 48 to 85%.

Therefore, rather than comparing the absolute concentrations in the buses vs. the taxis, the excess in-vehicle concentrations above the urban background are compared. This $PM_{2.5}$ level excess was not significantly higher (median test = 2.78, Fisher's exact p-value = 0.191) inside the buses ($29 \pm 10\ \mu g\,m^{-3}$) than in the taxis ($21 \pm 8\ \mu g\,m^{-3}$). The average Bus/Background and Taxi/Background ratios were 2.4 (range: 1.4–5.0) and 2.7 (range: 1.7–6.2), respectively.

Fig. 1 shows the scatter plots and linear regression models of $PM_{2.5}$ mass concentration in vehicles vs. the concurrent urban median concentrations by fixed-site monitoring stations. The linear regression equation describes the relationship between in-vehicle and ambient median $PM_{2.5}$ concentrations (b_1) and the average contribution of the in-traffic $PM_{2.5}$ sources (b_0). The regression model for buses was calculated excluding one bus outlier ($PM_{2.5} = 33.1\ \mu g\,m^{-3}$). The bus model is significant (F-test's p-value = 0.014; intercept (b_0) = 32 $\mu g\,m^{-3}$, 95% CI = 22–52 $\mu g\,m^{-3}$; slope (b_1) = 0.84, 95% CI = 0.2–1.49; $n = 14$). The taxi model is also significant (F-test's p-value = 0.0002; $b_0 = 20\ \mu g\,m^{-3}$, 95% CI = 11–29 $\mu g\,m^{-3}$; $b_1 = 1.01$, 95%, CI 0.59–1.44; $n = 14$).

The pDR $PM_{2.5}$ concentrations ranged from 24 to 96 $\mu g\,m^{-3}$ (M(SD) = 60(25) $\mu g\,m^{-3}$) and from 14 to 67 $\mu g\,m^{-3}$ (29(18) $\mu g\,m^{-3}$) inside the buses and the taxis, respectively (Table 2). Regarding the correlation between pDR and gravimetric $PM_{2.5}$ concentrations, only the pDR measurements obtained when the indoor RH% was below 60% were considered due to the bias associated with increased particle sizes when the RH% is higher than 60% (Chakrabarti et al., 2004). A good correlation between the

Table 2
Number of samples (n), arithmetic mean (M), standard deviation (SD), and range of gravimetric and pDR $PM_{2.5}$ mass concentrations and $PM_{2.5}$ filter absorption coefficient (Black Smoke Index, α) by type of vehicles

	Bus			Taxi		
	n	M (SD)	Range	n	M (SD)	Range
$PM_{2.5}$ mass concentration ($\mu g\,m^{-3}$)	15	56 (15)	33–77	14	39 (15)	20–70
pDR $PM_{2.5}$ concentration ($\mu g\,m^{-3}$)	11	60 (25)	24–96	10	29 (18)	14–67
Black Smoke Index: α ($m^{-1} \times 10^{-5}$)	15	11.5 (2.4)	6.1–14.7	14	11.5 (4.8)	3.8–18.1

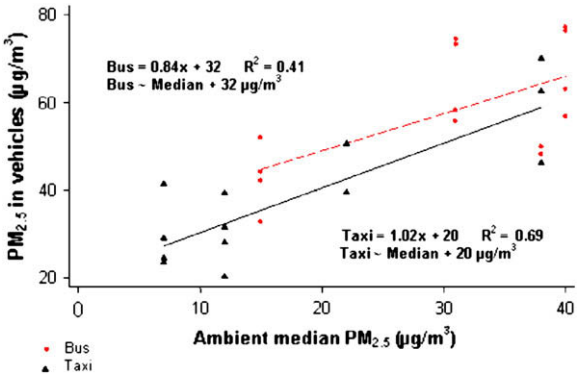

Fig. 1. Scatter plots and regression lines of $PM_{2.5}$ mass concentrations inside buses (dashed line) and inside taxis (full line) vs. ambient "urban median" 24 h-$PM_{2.5}$ mass concentrations.

pDR and the $PM_{2.5}$ mass concentrations ($R^2 = 0.68$) was observed inside the buses.

The BS index ranged from 6.1 to 12.7 $m^{-1} \times 10^{-5}$ and from 3.8 to 18.1 $m^{-1} \times 10^{-5}$ inside buses and taxis, respectively, with the same mean value of 11.5 $m^{-1} \times 10^{-5}$.

Table 3 displays the numbers of samples exceeding the MDLs, the Ms, the SDs, the 50th and 80th percentiles for those elements that exceeded the MDL in at least 80% of the samples, and only 50th and 80th percentiles for the other elements ($n = 3$–11). The mean elemental concentrations ranged from a few ng m^{-3} (V, Ni, and Br) to several µg m^{-3} (Fe, S). Sulfur is the predominant $PM_{2.5}$-bound element and ranged from 370 to 4380 ng m^{-3} inside buses, and from 380 to 2050 ng m^{-3} inside taxis. The elements detected in all bus and taxi samples were Cu, K, Si, S, Ca, Fe and Zn. For Br (MDL = 30 ng m^{-3}), Pb (MDL = 40 ng m^{-3}), and V (MDL = 20 ng m^{-3}), the MDLs were exceeded in only one or two of the 29 collected samples, and thus, no statistics are given in Table 3. The highest detected concentrations were Br = 35 ng m^{-3}, Pb = 55 ng m^{-3}, and V = 22 ng m^{-3}. With the exception of a few isolated samples, Pb and Br concentrations were below MDLs, reflecting the phasing out of lead from gasoline in the 1990s. In addition, V exceeded MDL in too few samples for any statistics.

Table 4 presents the Spearman's rank correlation coefficient (ρ_s) matrices for $PM_{2.5}$ mass and $PM_{2.5}$-bound element concentrations, and BS indexes for buses and taxis. Na, Mg, Ti, Mn and Ni exceeded the MDLs in less of 50% of the samples, and the respective data were considered insufficient for the correlation matrices. The values of $\rho_s \geq 0.8$ are shown in bold. There was significant correlation between the $PM_{2.5}$ mass and $PM_{2.5}$-bound element concentrations of Al, Si, S, K, and Zn and the BS index for the buses, as well as S and BS index for taxis.

Considering that the aerosol composition is less sensitive to meteorology than the absolute values of concentration; the percentages of the most abundant detected elements (on the weighed mass) by vehicle type have been calculated (Fig. 2). Clearly, the soil-related elements Al, Si, Ca, Fe, Cu and Zn contributed more to the $PM_{2.5}$ mass in the taxi samples, while the combustion-related S and K contributed more to the bus samples.

Assuming that all $PM_{2.5}$-bound S is in the form of diammonium sulfate, its contribution to the $PM_{2.5}$ mass was, on average, 19% (11 µg m^{-3}) and 11% (4 µg m^{-3}) in buses and taxis, respectively. The mass of the soil component, converting the Al, Si, Ca and Fe to their common oxides, resulted in about 12% and 19% of the $PM_{2.5}$ mass in the buses and taxis, respectively (corresponding to 6.5 µg m^{-3} for both).

Identification of the aerosol sources using receptor models was not possible due to the small number of samples (Henry et al., 1984). Multiple linear regression using BS index as a marker of traffic-related pollution, total S concentration as a marker of secondary aerosols, and Si concentration as a marker of the soil component explained 85% of the variability in $PM_{2.5}$ (R^2-adj = 0.85). The regression coefficients of the three variables were all significant, while the intercept value was comparable to zero (p-value = 0.9). The standard errors were 15% for both BS index and S regression coefficients, and errors higher than 35%

Table 3
MDL, number (n) of samples in which the specific element concentration was found above the MDL, arithmetic mean concentration (M), standard deviation (SD), 50th, and 80th percentiles of $PM_{2.5}$-bond elements for samples collected inside the buses and taxis (unit: ng m^{-3})

Species	Bus						Taxi					
	MDL	$n >$ MDL	M[a]	SD[a]	50th%	80th%	MDL	$n >$ MDL	M[a]	SD[a]	50th%	80th%
Al	63	15	230	109	210	300	86	13	200	115	245	299
Ca	42	15	820	321	720	1100	64	14	870	520	750	1450
Cl	70	3	[b]	[b]	109	160	97	4	[b]	[b]	156	180
Cu	7	15	52	22	52	63	10	14	85	48	74	115
Fe	6	15	1290	1020	893	2065	7	14	1250	1021	1245	1631
Mg	63	6	[b]	[b]	97	106	88	4	[b]	[b]	108	119
Mn	11	9	[b]	[b]	26	34	15	7	[b]	[b]	18	26
Ni	7	10	[b]	[b]	10	15	10	4	[b]	[b]	18	21
K	54	15	354	151	333	478	90	12[b]	160	100	147	213
Si	63	15	680	352	571	890	89	14	663	370	488	1105
Na	87	10	[b]	[b]	138	201	124	5	[b]	[b]	245	299
S	61	15	2720	1390	3009	3886	99	14	987	498	854	1238
Ti	30	5	[b]	[b]	41	46	44	1	[b]	[b]	[b]	[b]
Zn	8	15	54	15	53	65	10	14	55	17	53	72

[a] For M and SD calculations, MDL/2 values were used for all the cases below MDL.
[b] Statistics have been not calculated.

Table 4
Correlation matrices for elemental concentrations of the $PM_{2.5}$ aerosol (bus, taxi)

	Al	Si	S	K	Ca	Fe	Cu	Zn	BS
Bus $n = 15$ (Na through Pb)									
$PM_{2.5}$	0.55	0.58	**0.82**	**0.89**	0.22	0.46	0.42	0.51	0.67
Al		**0.93**	0.52	0.55	0.63	0.62	0.39	0.46	0.36
Si			0.59	0.58	0.73	0.68	0.35	0.36	0.31
S				**0.96**	0.32	0.50	0.36	0.25	0.28
K					0.32	0.46	0.39	0.34	0.43
Ca						0.45	0.19	0.33	0.30
Fe							**0.81**	0.20	0.27
Cu								0.29	0.42
Zn									0.55
Taxi $n = 14$ (Na through Pb)									
$PM_{2.5}$	−0.01	0.25	0.49	0.37	−0.11	−0.17	−0.30	0.03	**0.86**
Al		**0.85**	−0.10	0.48	**0.86**	0.46	0.34	0.23	−0.20
Si			0.04	0.49	**0.74**	0.41	0.30	0.24	−0.01
S				0.37	−0.20	−0.35	−0.15	−0.08	0.43
K					0.19	−0.10	−0.06	0.26	0.46
Ca						0.58	0.49	0.13	−0.35
Fe							**0.81**	0.71	−0.28
Cu								0.77	−0.29
Zn									0.15

Values denote the Spearman's rank coefficient of correlation (ρ_s). $\rho_s > 0.8$ are marked in bold.

affected the Si coefficient. The average contribution of the three sources was $59 \pm 8\%$ for the traffic component, $26 \pm 4\%$ for secondary aerosols, and $15 \pm 5\%$ for the soil component.

Each participant was engaged in some urban transporting, on average 2.4 h day^{-1} (8.5%), ranging from 0.5 to 5.2 h day^{-1} (2.1–21.9%). Table 5 shows the average time spent in the different means of commuting (%). According to the average time used, cars and walking are the most popular transportation means. More than of 60% of all subjects used a car for transport, on average, for 1.0 h day^{-1} (ranging from 0.1 to 5.3 h day^{-1}).

The personal 24-h average $PM_{2.5}$ exposure ranged from 6 to 56 $\mu g\,m^{-3}$, with a M(SD) of 20(11.9) $\mu g\,m^{-3}$. Meanwhile, the 24-h personal BS index ranged from 0.15 to 7.6 $m^{-1} \times 10^{-5}$ with an M (SD) of 3.4 (1.8) $m^{-1} \times 10^{-5}$.

Table 6 highlights the averages and ranges of the time spent on buses and in cars, the contributions of the exposures in these vehicles to $PM_{2.5}$ (and BS index) exposure

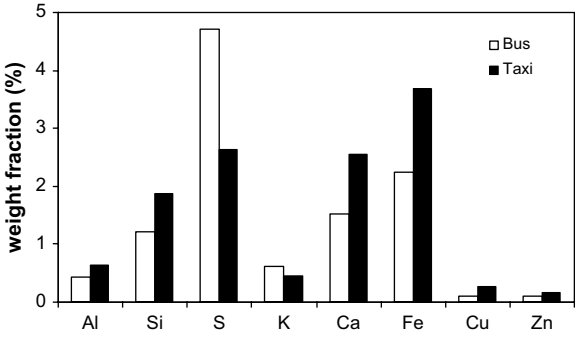

Fig. 2. Average contributions (%) of the most abundant and detected elements to the $PM_{2.5}$ mass concentration, for samples collected inside buses and taxis.

Table 5
Average daily time (range) spent in traffic in Florence

In-traffic	Users	% of time (range)
Walk	27/37	4.1 (0.5–15.6)
Bike	4/37	4.1 (2.1–6.3)
Motorcycle	8/37	6.4 (2.1–11.5)
Car/taxi	23/37	4.2 (0.35–21.9)
Bus	8/37	2.5 (0.5–5.2)
Total in traffic	37/37	9.4 (0.21–21.9)

Each percentage is for those who used the particular form of transport (%).

and the proportions of the total exposures acquired in them. On average, only 4.8% (68 min) of the total daily time is spent in cars and on buses. However, the average contribution of the time in these motor vehicles to $PM_{2.5}$ exposure is 2.5 times higher at 12%, and the average contribution to BS exposure is 5 times higher at 24%.

4. Discussion

The measured $PM_{2.5}$ concentrations on-board of the vehicles were well correlated with the contemporaneously 24-h $PM_{2.5}$ concentrations measured at fixed-site monitoring stations and with the "urban median". As expected from similar results obtained recently by other authors (Alm et al., 1999; Solomon et al., 2001; Fitz et al., 2003; Rodes et al., 1998; Wargo et al., 2002; Sabin et al., 2005), observed levels of $PM_{2.5}$ were higher than those measured at fixed-site monitoring stations. The in-vehicle $PM_{2.5}$ concentrations in excess of the "urban median" concentrations may be due to three origins: (i) near-field effects of non-traffic sources which concentrate in the vicinity of the busy streets, (ii) near-field effects of street dust suspension and tailpipe emissions of the other vehicles and their own exhaust on the same streets, and (iii) sources inside the vehicles, such as dust re-suspension, due to crowding and passenger activity.

The chosen buses and taxis traveled under the same typical high-traffic city conditions, where specific rush hours no longer exist because the traffic density remains almost at the same high level between 8 a.m. and 7 p.m.

Table 6
Ms (SDs) and ranges of the daily time spent, cumulative $PM_{2.5}$ and BS exposure in buses and cars, and the percent of the total exposure acquired in these vehicles for Florentine people ($n = 37$)

	Bus	Car	Bus + Car
	M (SD)	M (SD)	M (SD)
	Range	Range	Range
Average daily time spent in buses and cars (min)	8 (18) 0–75	60 (76) 0–315	68 (74) 0–315
Average daily cumulative $PM_{2.5}$ exposure in buses and cars ($\mu g \times m^{-3} \times$ min)	440 (1209) 0–4200	2334 (2946) 0–12 285	2773 (2960) 0–12 285
Average percent of $PM_{2.5}$ exposure acquired in buses and cars (%)	2 (6) 0–27	13 (21) 0–49	12 (14) 0–49
Average daily cumulative BS exposure in buses and cars ($m^{-1} \times 10^{-5} \times$ min)	86 (202) 0–825	688 (869) 0–3622	744 (860) 0–3622
Average percent of BS exposure acquired in buses and cars (%)	2 (6) 0–26	18 (30) 0–100	24 (34) 0–100

In this study, the $PM_{2.5}$ in-vehicle levels were about 29 $\mu g\,m^{-3}$ (bus) and 21 $\mu g\,m^{-3}$ (taxi) higher then the $PM_{2.5}$ urban background. The different $PM_{2.5}$ mass and elemental concentrations for taxis and buses may be due to different sampling periods but also other factors may have some influence as: (1) A bus was older, and two buses had inferior European emission standards than the taxis; (2) the sampled buses used ULSD while the taxis ($n = 3$) used CNG and/or gasoline that are clean fuels in relation to PM; (3) the buses were always packed in the city center, with consequently important PM re-suspension; (4) the buses run on fixed routes with predetermined authorized bus stops and can not escape from traffic jams so increasing idling; on the contrary the taxis cruise most of the operating time and have few stops; moreover the taxi drivers easily bypass the traffic congestions using alternative routes; (5) bus drivers, at the bus stops in the typical nose-to tail pattern, do not turn off engine but idling and when stationary open the three double-folding doors (span about 1.5 m) for get on and off the passengers.

In a study on pollution inside U.S.-school buses carried out in Connecticut, the Black Carbon (BC) and $PM_{2.5}$ levels inside the cabin were about 5–10 times higher than background concentrations (Wargo et al., 2002), with a clear increase of pollution when traveling behind buses (caravan) and heavy-duty diesel vehicles. Self-pollution, which occurs in buses when the bus's own exhaust migrates into the passenger cabin, was also found to play a significant role in on-board bus concentrations, especially when windows were closed (Solomon et al., 2001; Fitz et al., 2003; Sabin et al., 2005). The older buses showed higher rates of exhaust intrusion (Fitz et al., 2003; Behrentz et al., 2004). Hill et al. (2005) described two different ways for exhaust aerosol and gases to enter into the passenger compartment of a bus: (1) leaks from engine crankcase vent emissions (blow-by emission) and (2) exhaust plume traveling from the rear of the bus through the opened windows. The first phenomenon was discovered in U.S.-diesel-powered school buses (Borak and Sirianni, 2007). The orange Florentine buses have the engine under the floor and the exhaust pipe on the roof, and the crankcase emissions are re-circulated only for the Euro 3 or better emission standard-type buses.

In Wargo et al. (2002) study, on child personal exposure during a school day, the $PM_{2.5}$ were measured by real-time monitor/data logger pDR-1200, inside school buses without students under usual conditions of idling and driving on a rural route, during midday to minimize additional sources of PM. This study showed that in each bus run, $PM_{2.5}$ peak levels often occurred at, or near, stops due to exhaust and outdoor air entered inside through opened windows and doors and proved the effect of a moving bus, where $PM_{2.5}$ levels rapidly fell to background levels in case of open windows.

In our study, no information regarding bus ventilation was available. Nevertheless, we know that the majority of Florentine urban buses operate in high-traffic and narrow street canyons, have frequent stops and sometimes caravan.

Neither the BS index nor the elemental compositions was able to provide certain information regarding the proportion of $PM_{2.5}$ that might derive directly from primary (exhaust) traffic-related emissions. BS index is an optical measure of the carbonaceous chain agglomerates referred to as "soot", therefore, it is considered a good but non-specific marker for Diesel Exhaust Particles (DEPs) (Kinney et al., 2000; Adams et al., 2002; Health Effects Institute (HEI), 2003). The average BS indexes measured in the two types of vehicles were almost the same, as expected considering that they traveled on similar workdays. Particles originating from gasoline and diesel-powered engines are usually smaller than 300 nm in diameter (Wahlin et al., 2000). As a result, DEPs are virtually all found in particles with size less than 1 μm and, hence, are nearly completely (about 94%) contained within the $PM_{2.5}$ size fraction (Morawska et al., 1998; ARB, 1998; Gertler et al., 2002; Schneider et al., 2005).

Sulfur and potassium levels were higher inside buses than inside taxis, while copper concentration in buses was half of that in taxis. $PM_{2.5}$-bound S inside the bus cabin may be attributable (1) to regionally and long-range transported photochemical secondary sulfate and (2) to the infiltration of fresh local exhaust emissions from diesel engines containing primary sulfate.

The study by Reponen et al. (2003), conducted in the metropolitan residential area of Cincinnati (U.S.), reported a negative gradient of $PM_{2.5}$-bound S concentrations (up to 400 m) corresponding to the distance from highways, probably related to diesel engine emissions in areas where the traffic is the major local source of ambient air pollution.

A recent study showed a higher sulfur level in diesel-exhaust-$PM_{2.5}$ collected in vehicles with frequent idling than the concentrations obtained in similar vehicles traveling with a constant speed of 40 $km\,h^{-1}$ (Saitoh et al., 2003). Moreover, sulfate was found to be comprised from 1 to 14% of diesel particle mass, depending on fuel composition (HEI, 2002; ARB, 1998). In all diesel related averages, between 1.0 and 2.1% of the $PM_{2.5}$ mass was sulfate (Schauer et al., 2006). However, based on the low sulfur ($<$10 ppm) content of the diesel used in Florence and because there is no significant difference in the BS index between the bus and taxi results, the higher sulfur levels in the buses are unlikely to be caused by self-pollution but likely from outdoor pollution. Sulfate particles have a long persistency time in the ambient air; therefore, it is reasonable that local and micro-scale sources (principal diesel-derivate particles in this specific case-study because the domestic heating systems were off during sampling period and there are not local coal industrial sources), as well as slowly varying regional background, all contribute to the overall sulfur concentration. $PM_{2.5}$-bound S data were not available for any urban fixed-site monitoring stations on exactly the same days as bus and taxi data, so they could not be compared directly to positively establish a $PM_{2.5}$-bound S excess on-board of buses and taxis.

A previous Florentine $PM_{2.5}$ elemental speciation study carried out in 2001 at Gramsci Street (traffic site) gave average $PM_{2.5}$-bound S concentrations of 1282 ± 524 and 1372 ± 462 $ng\,m^{-3}$ in the winter and the summer, respectively (D'Alessandro et al., 2003, 2004; Ariola et al., 2002). Compared to these data, the range of $PM_{2.5}$-bound S concentrations (370–4380 $ng\,m^{-3}$) measured on-board of

the buses in the fall was quite high but still in the range of variation of the previous studies.

Slightly different patterns of correlation between certain elements are described in Table 4: good correlation is seen for S, K, and $PM_{2.5}$ concentrations in the bus and for the $PM_{2.5}$ mass concentration and BS index in taxis. The trace metal concentrations do not correlate well with the $PM_{2.5}$ concentration because of their small contribution to the total mass.

Today, no information about $PM_{2.5}$ self-pollution inside diesel-powered bus cabins is available in Italy. Despite the limited number of $PM_{2.5}$ data on-board vehicles, these give the first insight into the Florentines' exposure concentrations when commuting. The samples were collected during traffic polluted workdays and under real operative conditions, when summer holidays were already finished, schools were opened, and domestic heating was not yet operating (operating from November, 1st) and what's more the vehicle data were not affected by smoking pollution.

It is not possible to establish if there were different $PM_{2.5}$ levels in front and rear part of the bus, nevertheless even if the PEM was placed between the front entrance and central exit doors, there were no partitions inside the bus cabin and usually the passengers were moving towards the exit, then we consider that the measured levels may represent acceptable data for the exposure estimation of the users.

The assessment results suggested that the user's exposure encountered on-board of buses and/or cars during commuting or riding is of greater importance due long travel time spend per day and the high peak pollution levels. The other traffic-related exposures such us that occur at bus loading/unloading zones or during waiting in areas near bus stops, trunk roads, junctions, and other vehicle emission polluted areas, due to the short lengths of dwell or wait time, could not contributes significantly to people daily and annual exposures to $PM_{2.5}$ especially if these activities are sporadic.

In conclusion, these results are of particular concern: even if the commuting time on-board of vehicles is a small fraction of an individual's day (except for professional drivers), it appears important to minimize the Florentine citizen's exposure to $PM_{2.5}$ while commuting. Furthermore, considering the strong emphasis at different levels to increase bus use, in an effort to reduce urban traffic congestion and air pollution, it would be wise to introduce more priority bus-only lanes and replace the oldest buses in the fleet with new less-polluting ones, to install pollution control devices like oxidation catalysts and particulate traps, to reduce bus "caravanning" and cut down the unnecessary idling time, to use alternative fuels to diesel such as gaseous fuels (CNG, LPG). These steps would help to reduce the personal in-traffic exposure to $PM_{2.5}$ of the bus and car riders.

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