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A comprehensive assessment of PM emissions from paved roads: Real-world Emission Factors and intense street cleaning trials

F. Amato [a,*], S. Nava [b], F. Lucarelli [b], X. Querol [a], A. Alastuey [a], J.M. Baldasano [c,d], M. Pandolfi [a]

[a] Institute of Environmental Assessment and Water Research, Spanish Research Council (IDÆA-CSIC), c/Jordi Girona 18-26, 08034 Barcelona, Spain
[b] National Institute of Nuclear Physics (INFN) and Physics Department, University of Florence, via Sansone 1, 50019 Sesto Fiorentino, Italy
[c] Environmental Modelling Laboratory, Technical University of Catalonia, Avda. Diagonal 647, Edificio H, Oficina 10.23, 08028 Barcelona, Spain
[d] Barcelona Supercomputing Center, Centro Nacional de Supercomputación (BSC-CNS), Earth Sciences Department, Jordi Girona 29, Edificio Nexus II, 08034 Barcelona, Spain

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ABSTRACT

Compliance with air quality standards requires control of source emissions: fine exhaust particles are already subject to regulation but vehicle fleets increase whilst the non-exhaust emissions are totally uncontrolled. Emission inventories are scarce despite their suitability for researchers and regulating agencies for managing air quality and PM reduction measures. Only few countries in Europe proposed street cleaning as a possible control measure, but its effectiveness is still far to be determined.

This study offers first estimates of Real-world Emission Factors for PM_{10} and brake-wear elements and the effect on PM_{10} concentrations induced by intense street cleaning trials.

A straightforward campaign was carried out in the city of Barcelona with hourly elemental composition of fine and coarse PM to detect any short-term effect of street cleaning on specific tracers of non-exhaust emissions. Samples were analyzed by Particle Induced X-Ray Emission.

Real-world Emission Factor for PM_{10} averaged for the local fleet resulted to be 97 mg veh^{-1} km^{-1}. When compared to other European studies, our EF resulted higher than what found in UK, Germany, Switzerland and Austria but lower than Scandinavian countries. For brake-related elements, total EFs were estimated, accounting for the sum of direct and resuspension emissions, in 7400, 486, 106 and 86 μg veh^{-1} km^{-1}, respectively for Fe, Cu, Sn and Sb. In $PM_{2.5}$Fe and Cu emission factors were respectively 4884 and 306 μg veh^{-1} km^{-1}.

Intense street cleaning trials evidenced a PM_{10} reduction at kerbside of 3 μg m^{-3} (mean daily levels of 54 μg m^{-3}), with respect to reference stations. It is important to remark that such benefit could only be detected in small time-integration periods (12:00–18:00) since in daily values this benefit was not noticed. Hourly PM elemental monitoring allowed the identification of mineral and brake-related metallic particles as those responsible of the PM_{10} reduction.

1. Introduction

Several adverse health effects have been associated to exposure to atmospheric particulate matter (PM). Long term exposure to high concentrations of PM increases the risk of lung cancer, respiratory disease and arteriosclerosis, whereas short-term exposure peaks can cause exacerbation of several forms of respiratory diseases and changes in heart rate variability (Chiaverini, 2002; Samet et al., 2000). Brunekreef and Forsberg (2005) highlighted the significant health burden of coarse particles (between 2.5 and 10 μm). They concluded that there is some evidence for effects of coarse PM on mortality and this is more prominent in studies from arid regions. In urban environments coarse particles are mainly controlled by anthropogenic mineral dust emissions. Toxicological studies found that, on a mass basis, cytokine production and inflammatory response of macrophages were especially enhanced by coarse PM (Becker et al., 2002). Moreover transition metals, such as Co, Cu, Fe, Mn, Ni, Ti and V contribute to the oxidative capacity of PM (Prahalad et al., 1999; Clarke et al., 2000) and Cr, Mn and Fe the main determinants of PM-induced cytotoxicity.

However, since no definitive threshold for PM-induced adverse health effects has been yet established, there is a general consensus that ambient PM levels should be reduced as much as possible. Given that road traffic is a major source of urban PM and atmospheric metals, air quality managers are recently focusing on this sector for specific emission control measures. Road traffic emissions comprise not only tailpipe exhaust emissions but also non-exhaust emissions derived mostly from the vehicle-induced resuspension of dust deposited on the road and in part from the direct emissions from vehicle wear (brakes, tires, discs etc.). Non-exhaust emissions are often of the same order or even greater than exhaust emissions, especially when rainfall rates are low and the wash-off of the road is

* Corresponding author. Tel.: +34 934095410; fax: +34 93 411 00 12.
E-mail address: fulvio.amato@idaea.csic.es (F. Amato).

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reduced (Amato et al., 2009a; Kousoulidou et al., 2008; Kristensson et al., 2004; Abu-Allaban et al., 2003).

Despite the high environmental and health burden of non-exhaust sources in urban environments, their emissions are often unknown and still uncontrolled. In the Iberian Peninsula for example, no Real-world Emission Factors (EFs) are currently available for researchers and regulating agencies for managing air quality and PM reduction measures. Moreover, there is still a dearth of knowledge on the effectiveness of some possible remediation measures such as street cleaning activities. Street cleaning can be performed by sweeper vehicles and/or water jet cleaning. In their recent review of efficiency tests, Amato et al. (2010), concluded that the street sweeping alone did not show any benefit in the short term (Kuhns et al., 2003; Gertler et al., 2006; Kantamaneni et al., 1996; Norman and Johansson, 2006), while more encouraging results have been achieved so far with the combined use of sweepers and water jet cleaning. Nevertheless, the whole effectiveness of these procedures is still far from being definitively determined since only few tests have been carried out so far and with very different local conditions. Also energy and water consumptions are of some concern. These can be reduced by means of the use of electric/hybrid sweepers and the use of no-drinking groundwater. Given this uncertainty more field studies are needed and additional tools, apart from PM monitoring, should be used to further investigate the effectiveness of street cleaning, minimizing the effect of meteorology, and to quantify the PM benefit in the vicinity of the road.

The present study offers new insights from a comprehensive field study aimed to estimate Real-world EFs and to evaluate the impact of street cleaning activities in ambient concentration of PM and non-exhaust emissions. An intensive campaign was carried out during spring 2009 in the city centre of Barcelona (NE of Spain) by means of the application of innovative techniques: i) the hourly elemental composition of size segregated PM was used to investigate short-term variability of specific tracers of road dust resuspension, in order to detect variations in road dust emissions within a single day; ii) a Positive Matrix Factorization was applied in order to identify the daily pattern of each PM source; iii) Real-world Emission Factors (EFs) for PM_{10} resuspension and brake-wear tracers were estimated and expressed in terms of micrograms per vehicle per kilometer travelled. To our knowledge, this approach has been never applied in order to evaluate the effectiveness of street cleaning.

The city of Barcelona represents somehow a typical Mediterranean scenario concerning road traffic density and weather conditions. In the city centre passenger car density is of one of the highest in Europe (6100 veh km^{-2}) and precipitations are scarce and infrequent. Consequently, with respect to the central European countries, the humidity of road surface is lower and leaves to higher road dust emissions. Previous source apportionment studies reveal that PM_{10} pollution in Barcelona is mainly driven by local sources: road traffic exhaust (21%), non-exhaust (17%) and construction sites (25%) (Amato et al., 2009a). This is reflected by the daily exceedances of EU limit values, which in 2008 were more than 35 for all the monitoring sites located at busy roads (5) and for half (2 out of 4) of the urban background monitoring sites (ASPB, 2009), while no one of the mentioned air quality stations went beyond the number of exceedances (18) permitted for NO_2. Such scenario is mainly due to the construction works (about 300 within the city in 2008) and to the consequent widespread road dust emissions.

2. Methodology

The street cleaning effectiveness was tested in a busy road of the city centre of Barcelona: *Valencia Av.* (19,000 veh day^{-1}). In the city centre traffic is dominated by passenger cars (63%) and motorcycles (27%) with a small proportion of light and heavy duty vehicles (7%) (http://w3.bcn.es/fitxers/mobilitat/dadesbasiques2006.222.pdf). Traffic flow is horizontal, unidirectional (WSW–ENE) from the city centre heading to the northern exit of the city. *Valencia Av.* is a 19 m wide five-lane road, including a left lane reserved for parking lots. Building height varies between 6 and 7 storeys, making *Valencia Av.* a street canyon with secondary roads intersecting it perpendicularly in octagonal crossings. A traffic counter was available by the Municipal Transport Service measuring total number of vehicles every 15 min.

Air quality measurements were performed during 5 weeks (February–March 2009) being the street cleaning (StC hereafter) carried out only during the third week, every night from 23:00 to 02:00 approximately. StC was performed into a 1200 m long stretch of *Valencia Av.* by the urban cleaning agency BCNeta (vacuum assisted mechanical sweeper vehicle: RosRoca Citycat 5000 SL/XL), followed by manual (hoses) groundwater jet, resulting in a street run-off of approximately 0.3 l m^{-2} comprising sidewalks, active lanes and parking lane.

Three mobile laboratory vans were used for this study. Two vans (VAL1 and VAL2) were installed along *Valencia Av.* at a distance of approximately 1200 m one from each other and at the extreme points of the cleaned stretch. A third van (COR) at *Corsega Av.*, a less busy road (11,000 veh day^{-1}) parallel to *Valencia Av.* but enough far away to represent a reference station. All the three vans were parked on the left lane. Seven further stations belonging to the air quality network were used as reference sites, comprising 4 traffic sites and 3 urban background sites. At these stations PM_{10} was sampled with different frequencies, therefore different comparisons were made, one for each site. PM_{10} sampling was carried out in 24 h cycles (22:00–22:00) by DIGITEL and MCV sequential high volume samplers (30 m^3 h^{-1}). Light scattering particle counters GRIMM (1.108 and 1.107) measured levels of PM_{10} every 30 min and average daily concentrations were corrected by inter-comparison with gravimetric data. Conventional analyzers were used for measurements of levels of gaseous pollutants. Meteorological towers were installed for wind speed and direction (4 m height).

About a hundred PM_{10} samples were collected onto quartz fiber filters (Ø 15 cm, Munktell) and analyzed following the procedures described by Querol et al. (2001) to determine concentration of Al, Ca, K, Mg, Fe, Ti, Mn, P, S, Na, NO_3^-, Cl^-, NH_4^+ and 23 trace elements. Uncertainties were calculated according to the methodology described in Amato et al. (2009a).

Hourly elemental concentrations were obtained at VAL1 by a two-stage (coarse and fine PM) STREAKER sampler. Full details of the sampler, its cut-off diameters, control unit, etc. can be found elsewhere (Formenti et al., 1996). Three pairs of stages (3 weeks: from 9th to 27th March) were collected and analyzed by Particle Induced X-Ray Emission (PIXE) at the LABEC-INFN facility in Florence (based on a 3 MV Tandetron accelerator, where an external beam set-up is fully dedicated to atmospheric aerosol studies (Calzolai et al., 2006; Chiari et al., 2005)). X-ray spectra have been fitted for 25 elements (Na, Mg, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, As, Se, Br, Rb, Sr, Zr, Mo and Pb) using the GUPIX software package (Maxwell et al., 1995) and the elemental concentrations were obtained via a calibration came from a set of thin standards certified within 5% (Micromatter Inc.).

A Positive Matrix Factorization (PMF) model was applied to the chemical dataset (349 samples) obtained through the PIXE analysis, in order to investigate the temporal variability of the main PM sources, both in a daily scale and also along the campaign. To this aim the Multilinear Engine (Paatero, 1999) scripting language was used to create a specific PMF problem. Concentrations of species in the fine and coarse fractions were summed in order to create a PM_{10} database. Data uncertainties σ_{ij} were calculated as:

$$\begin{aligned} \sigma_{ij} &= \sigma_{ij}^{a} + 0.05 \cdot x_{ij} \qquad \text{if } x_{ij} > DL_i \\ \sigma_{ij} &= \frac{3}{2} \cdot DL_i \qquad \text{if } x_{ij} \le DL_i \end{aligned} \tag{1}$$

where x_{ij} is the concentration of the ith species at the jth hour, σ_{ij}^{a} is the analytical error calculated for the ith species at the jth hour by the

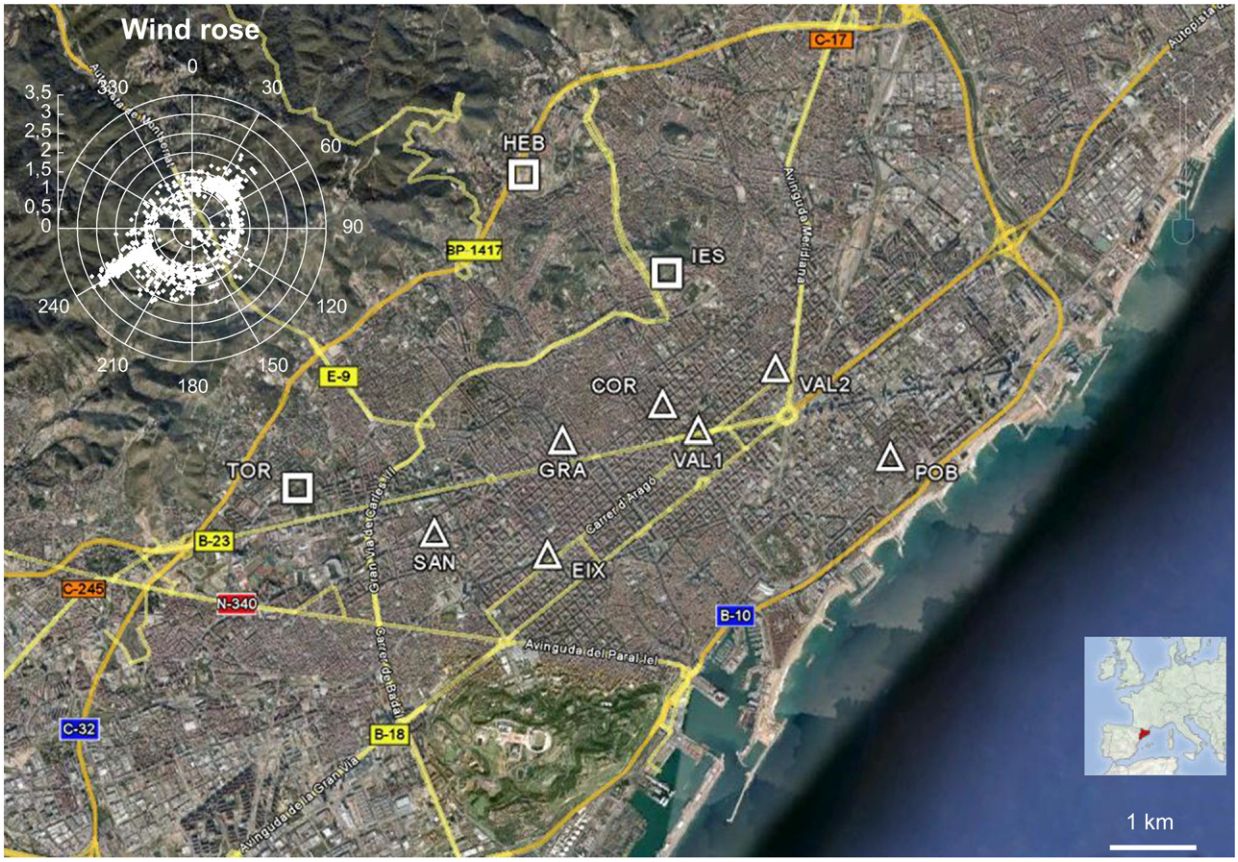

Fig. 1. Study area (main traffic routes are highlighted), location of VAL1, VAL2 and COR laboratory vans, seven reference air quality stations and wind speed (m/s) and direction. Triangles indicate traffic sites, squares indicate urban background sites.

GUPIX software which includes the counting statistic and the fit uncertainty and DL_i are the detection limits calculated for each species calculated following the three-sigma criterion. Basing on the signal to noise criterion and also on the percentage of data above the detection limit, only 21 species were selected as strong species.

For the calculation of emission factors a linear model was used, basing on the two assumptions mentioned already in the Introduction section. More precisely, at *Valencia Av.* the ambient concentrations of one pollutant come from the sum of different contributions: i) an urban background; ii) the local traffic source; iii) other possible local sources. At *Corsega Av.* the same is valid but the local traffic source is significantly lower, given that traffic volume is almost 50% less. Our approach was the search of a linear regression between the increments of road traffic emissions between *Valencia Av.* and *Corsega Av.*, and the increment of PM_{10} concentrations. The average fleet emission factor for the local fleet ($EF_{x,\ fleet}$) was estimated from the fitting of the following equations:

$$\Delta x_j = \frac{EF_{x,fleet} \cdot \Delta n_j}{d} + C \tag{2}$$

where Δx_j is the concentration difference of the pollutant x on the jth 24-hour sample between *Valencia Av.* (average of VAL1 and VAL2) and *Corsega Av.* (COR), Δn_j is the difference of total number of vehicles, C is a fitting constant accounting for other possible local sources and d is the factor accounting for the atmospheric dilution for traffic related emissions, obtained by:

$$d = \frac{EF_{NOx} \cdot \Delta n_j}{\Delta NOx} \tag{3}$$

where, EF_{NOx} are reliable emission factors for NOx during working and weekend days, calculated for the local fleet composition, speed data and working/weekend days according to the methodology of Baldasano et al. (2008) who used the CORINAIR-Computer Programme to Calculate Emissions from Road Transport (COPERT III, Ntziachristos and Samaras, 2000) and ΔNOx is the concentration difference of NOx.

Table 1
Mean values of PM10 gravimetric concentrations during StC days and no-StC days at the intervention site and at reference sites.

Common weekdays	Cleaned road sites		Traffic reference sites					Urban background reference sites		
	VAL1	VAL2	COR	EIX	GRA	POB	SAN	IES	HEB	TOR
StC	66	71	58							
No StC	54	54	49							
Increment	12	17	9							
StC	66	71		64						
No StC	52	51		44						
Increment	14	19		20						
StC	66	71			54					
No StC	52	52			41					
Increment	14	18			13					
StC	66	71				72				
No StC	52	52				49				
Increment	14	18				24				
StC	66	71					69			
No StC	52	52					51			
Increment	13	18					18			
StC	66	71						56		
No StC	52	52						34		
Increment	14	19						22		
StC	66	71							61	
No StC	54	55							42	
Increment	12	16							19	
StC	66	71								48
No StC	54	54								41
Increment	12	17								7
Average increment	13 ± 1	18 ± 1					16 ± 6			

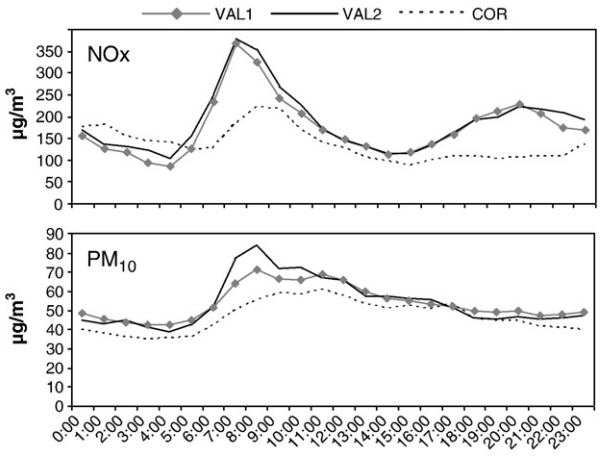

Fig. 2. Average daily evolution of NOx and PM_{10} concentrations at VAL1, VAL2 and COR.

Similarly to previous studies (Bukowiecki et al., 2009; Johansson et al., 2009) we used Eq. (3) basing on the assumption that the dilution of the vehicle related emissions from the point of emission to the sampler inlets was comparable for NOx and the particulate pollutants. Only daily values with $\Delta NOx > 20\ \mu g\ m^{-3}$ were selected. This approach has been used in several earlier studies (Ketzel et al., 2003; Omstedt et al., 2005; Johansson et al., 2009), showing a satisfactory theoretical basis. This is basically based into two assumptions: a) emissions affecting the reference site are affecting also the more trafficked site; b) emissions at the trafficked site have a negligible impact into the reference site. In the present case a linear regression was investigated for PM_{10} and for several species, typically related to brake wear, such as Fe, Cu, Zn, Sb, Ba, Sn, Mo and Zr. However for the EFs calculation we stated as minimal requirement that at least a fair fit between the couples of Δx_j and $\Delta NOx_j/EF_{NOx}$ values was obtained. The fleet emission factor is valid for the average traffic composition during the considered time period and includes contributions from direct (not previously deposited) and resuspended particles.

Table 2
Comparison between mean concentrations ± standard deviation of PM10, major ($\mu g\ m^{-3}$) and trace components ($ng\ m^{-3}$) obtained from 24-hour accumulated particles by means of high volume samplers (no rainfall days); In: inorganic aerosol, el: elements, Und: undetermined, DL: detection limit.

	StC weekdays			No-StC weekdays		
$\mu g\ m^{-3}$	VAL1 upwind	VAL2 downwind	COR reference	VAL1 upwind	VAL2 downwind	COR reference
PM_{10}	66 ± 14	71 ± 10	58 ± 11	56 ± 17	56 ± 20	50 ± 18
OC	6.4 ± 1.5	6.7 ± 1.3	6.9 ± 1.2	5.3 ± 1.9	5.7 ± 2.4	5.2 ± 2.1
EC	6.7 ± 1.3	8.2 ± 1.2	5.0 ± 1.3	5.3 ± 1.1	5.2 ± 1.7	3.6 ± 1.3
Al	0.7 ± 0.2	1.0 ± 0.2	0.7 ± 0.2	0.6 ± 0.3	0.7 ± 0.4	0.6 ± 0.3
Ca	3.0 ± 1.2	4.3 ± 1.1	2.6 ± 1.1	2.3 ± 1.0	3.0 ± 1.2	2.1 ± 1.0
Fe	2.6 ± 0.6	2.8 ± 0.3	1.8 ± 0.4	2.3 ± 0.5	2.0 ± 0.6	1.4 ± 0.5
K	0.5 ± 0.1	0.6 ± 0.1	0.5 ± 0.1	0.4 ± 0.2	0.4 ± 0.2	0.4 ± 0.2
Mg	0.3 ± 0.1	0.4 ± 0.1	0.3 ± 0.2	0.3 ± 0.1	0.4 ± 0.2	0.4 ± 0.1
Na	1.0 ± 0.6	1.0 ± 0.7	0.9 ± 0.6	1.4 ± 0.7	1.4 ± 0.7	1.5 ± 0.8
P	0.1 ± 0.0	0.1 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0
S	1.4 ± 0.3	1.7 ± 0.3	1.6 ± 0.2	1.0 ± 0.5	1.1 ± 0.5	1.2 ± 0.5
Cl^-	0.8 ± 0.9	2.6 ± 0.9	1.0 ± 0.7	1.3 ± 0.7	2.3 ± 0.9	1.5 ± 0.9
NO_3^-	6.4 ± 0.7	5.5 ± 1.0	6.7 ± 1.2	5.8 ± 3.7	5.5 ± 3.4	6.6 ± 4.2
$SO_4^=$	3.4 ± 0.7	2.8 ± 0.5	3.9 ± 0.9	2.5 ± 1.2	2.5 ± 1.3	3.1 ± 1.5
NH_4^+	1.6 ± 0.2	1.9 ± 0.2	1.8 ± 0.2	1.2 ± 0.8	1.4 ± 1.1	1.6 ± 1.1
$ng\ m^{-3}$						
Li	0.8 ± 0.3	1.0 ± 0.2	0.7 ± 0.2	0.7 ± 0.3	0.8 ± 0.4	0.6 ± 0.3
Ti	62.4 ± 18.2	72.0 ± 18.2	49.6 ± 13.9	51.7 ± 19.8	48.1 ± 22.6	42.3 ± 18.3
V	17.0 ± 3.5	19.9 ± 5.6	17.4 ± 3.4	9.6 ± 5.2	12.1 ± 6.9	11.0 ± 6.1
Cr	20.6 ± 4.5	26.5 ± 8.9	22.1 ± 3.4	19.1 ± 5.2	15.0 ± 4.8	13.0 ± 5.2
Mn	36.1 ± 10.0	39.6 ± 6.5	26.3 ± 7.3	32.1 ± 9.0	29.5 ± 9.3	23.6 ± 7.4
Co	0.5 ± 0.1	0.8 ± 0.2	0.5 ± 0.1	0.4 ± 0.1	0.5 ± 0.2	0.4 ± 0.1
Ni	7.4 ± 2.0	9.1 ± 2.2	8.3 ± 1.6	5.0 ± 2.2	6.6 ± 3.1	5.6 ± 2.6
Cu	137.9 ± 28.1	121.0 ± 13.0	80.4 ± 12.6	119.0 ± 25.5	88.9 ± 25.0	61.7 ± 22.7
Zn	104.1 ± 29.6	108.6 ± 19.7	80.6 ± 28.0	100.0 ± 38.8	89.0 ± 29.2	88.5 ± 39.8
As	0.9 ± 0.2	1.0 ± 0.2	0.8 ± 0.2	0.8 ± 0.3	0.8 ± 0.3	0.7 ± 0.3
Se	1.4 ± 0.3	1.5 ± 0.3	1.4 ± 0.3	1.1 ± 0.7	1.2 ± 0.6	1.2 ± 0.7
Rb	1.5 ± 0.4	1.8 ± 0.4	1.3 ± 0.4	1.3 ± 0.6	1.3 ± 0.7	1.1 ± 0.6
Sr	7.4 ± 2.3	9.2 ± 2.1	6.2 ± 1.8	6.6 ± 2.3	7.6 ± 3.2	6.1 ± 2.7
Zr	17.2 ± 2.1	17.9 ± 1.1	16.1 ± 1.7	16.0 ± 2.1	15.3 ± 2.2	12.9 ± 4.1
Mo	10.8 ± 1.6	11.1 ± 7.0	6.3 ± 1.2	8.3 ± 2.8	7.9 ± 6.9	4.0 ± 2.6
Cd	0.5 ± 0.3	0.7 ± 0.5	0.5 ± 0.2	0.3 ± 0.2	0.5 ± 0.6	0.4 ± 0.4
Sn	28.8 ± 3.4	22.7 ± 1.8	17.4 ± 1.0	24.6 ± 5.5	17.1 ± 5.6	12.9 ± 5.1
Sb	17.9 ± 4.2	16.6 ± 2.0	11.9 ± 3.3	15.0 ± 3.7	11.4 ± 3.8	7.6 ± 3.2
Cs	0.11 ± 0.0	0.12 ± 0.0	0.11 ± 0.0	0.09 ± 0.0	0.10 ± 0.1	<DL
Ba	76.0 ± 12.8	51.8 ± 8.7	28.4 ± 10.0	57.2 ± 16.8	32.8 ± 12.8	22.2 ± 14.0
La	0.5 ± 0.1	0.6 ± 0.1	0.5 ± 0.1	0.5 ± 0.2	0.5 ± 0.3	0.5 ± 0.2
Ce	1.7 ± 0.5	1.7 ± 0.3	1.5 ± 0.4	1.4 ± 0.5	1.3 ± 0.5	1.2 ± 0.5
Hf	0.7 ± 0.1	0.7 ± 0.0	0.7 ± 0.5	0.6 ± 0.1	0.6 ± 0.1	0.5 ± 0.2
Pb	14.3 ± 4.1	14.2 ± 0.0	13.5 ± 4.4	12.8 ± 5.2	11.0 ± 3.5	12.6 ± 5.7
$\mu g\ m^{-3}$						
Mineral	16.0	22.2	14.0	13.1	15.7	12.2
Secondary In.	11.3	10.1	12.4	9.5	9.5	11.4
Sea salt	1.7	3.6	1.9	2.8	3.7	3.1
Trace el.	0.5	0.4	0.3	0.4	0.3	0.3
Und. (%)	35	27	30	35	28	29

3. Results

3.1. General conditions

The last important rainfall (>0.2 mm) before the starting of the measurement campaign occurred between 31st January and 3rd February (52 mm), although during the campaign there were other sporadic rainfall episodes during the last week. Wind direction (measured 4 m above ground level) was typically an alternate SW–NE channeled flow along *Valencia Av.*, with a predominant SW direction due to the sea-breeze development from 10:00 to 17:00 h typically. A negligible cross-street component was likely due to the turbulence generated by vehicles flow (Fig. 1). Wind speed was light varying within 1–2 m/s during night with sea-breeze peaks up to 3 m/s between 11:00 and 16:00. VAL2 laboratory van was therefore predominantly downwind respect to VAL1 van. Traffic was typically 19,000 vehicles per weekday, with the highest densities occurring between 08 and 20 h local time (1200 veh h^{-1}).

The possible atmospheric long range transport conditions were investigated by calculating the daily movement pathways of air masses into the studied area by means of the HYSPLIT model 4 with vertically modeled transport back-trajectories. These interpretations were also coupled with information obtained from NAAPS aerosol dust concentration maps. During the campaign two main atmospheric conditions were observed, namely Atlantic advection and regional pollution episodes. The Atlantic advection episodes are characterized by the renewal of the air masses with subsequent reduction in the ambient air levels of PM whilst regional episodes are characterized by low pressure gradient and low circulation with consequent atmospheric stagnation. Days in which street cleaning was performed coincided with regional pollution episodes and pollutants stagnation and build up, consequently, at the reference sites PM_{10} registered mean increases within 17–65% (Table 1) and 34% increments in NOx at the reference COR site.

3.2. Emission factors

The daily increments of all analyzed species were investigated in their relationship with increments of $\Delta NOx_j/EF_{NOx}$. In Fig. 2 the average daily evolutions of NOx and PM_{10} are represented, revealing that atmospheric pollution increases sensibly at *Valencia Av.*, with respect to *Corsega Av.*, at two moments of the day: the rush morning hours (07:00–09:00, for both NOx and PM_{10}) and an evening peak (20:00) only for NOx. For brake-wear related elements and the rest of PM_{10} components, the average increments can be deduced from Table 2.

For the EFs calculation, a basic requirement in this study was a satisfactory linear fit for the considered pollutant. Among the investigated pollutants, only PM_{10}, Fe, Cu, Sb and Sn accomplished this requirement (Fig. 3). The rest of elements (Zn, Mo, Ba, Pb and Zr) did not show any appreciable relationship between the increase of traffic emissions ($\Delta NOx_j/EF_{NOx}$) and the increase in their ambient concentrations (Δx). This can be due to several reasons: i) these species are not specifically tracers of traffic emissions; ii) the uncertainty of measurements did not permit to observe a linear relationship. Thus, even if EFs could be calculated day by day (without regression), we decided not to present EFs results for these elements.

For PM_{10}, the total EF was fitted in 0.158 g veh^{-1} km^{-1}, this including both exhaust and non-exhaust emissions. The EF only for non-exhaust was estimated by subtracting a representative average exhaust EF for the local fleet (Baldasano et al., 2008). The EF_{res} (for resuspension) resulted in 97 mg veh^{-1} km^{-1}, rather higher than EF_{exh} (61 mg veh^{-1} km^{-1}). This result is in agreement with previous studies (Westerlund, 2001 as an example) and a recently published source apportionment study in Barcelona, indicating that average contributions from non-exhaust sources were similar to those from exhaust on an annual basis (Amato et al., 2009a). The fitting constant *C* (Eq. (2)) was found to be negligible revealing that traffic was nearly the unique source responsible of the PM_{10} increments between *Corsega Av.* and *Valencia Av.* When compared to other European studies, our EF_{res} resulted higher than mixed fleet EFs found in UK (14–23 mg veh^{-1} km^{-1}, Thorpe et al., 2007), Germany (57–67 mg veh^{-1} km^{-1}, Ketzel et al., 2007), Switzerland (27 mg veh^{-1} km^{-1}, Bukowiecki et al., 2010; see also Gehrig et al., 2004) and Austria (62 mg veh^{-1} km^{-1} for the sum of exhaust and non-exhaust sources by Handler et al., 2008) but lower than in Scandinavian countries (Kristensson et al., 2004; Sternbeck et al., 2002; Ketzel et al., 2007). An exhaustive review is available from Schaap et al. (2009).

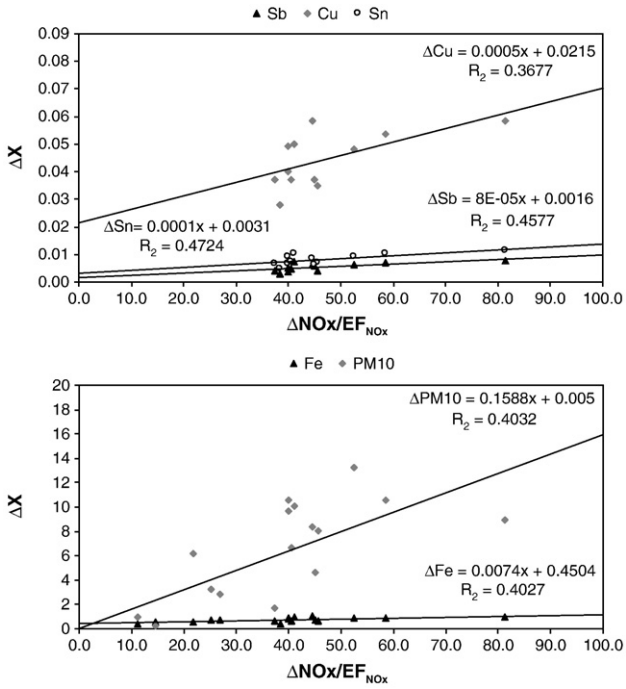

Fig. 3. Emission factor estimates for PM10, Fe, Cu, Sn and Sb, by means of regression between increments in pollutant concentrations (*y* axis, μg m^{-3}) and increments in normalized NOx concentrations (*x* axis).

As shown in Fig. 3, Sn and Sb showed the best fits revealing EF of 106 μg veh^{-1} km^{-1} and 86 μg veh^{-1} km^{-1}, respectively. These EFs account for both the direct and resuspension emissions. A comparison with literature EFs is provided in Table 3. The *C* constant was in these cases relatively (in %) more significant, confirming that the fraction of ΔPM due to non traffic sources and non local traffic emissions was in the order of ng m^{-3}. These values, similar to what obtained in previous studies (see Bukowiecki et al., 2009 as example) as well as those for r^2, were robust enough to make reliable the EFs calculation. For Fe and Cu the fitted EF were respectively 7400 μg veh^{-1} km^{-1} and 486 μg veh^{-1} km^{-1}. The unexplained fraction rose at 18–20% for these two elements but revealing still main emissions from road traffic. When coupling such results with the size segregated concentrations obtained by the STREAKER we estimated that for $PM_{2.5}$ the EFs were 4884 and 306 μg veh^{-1} km^{-1} for Fe and Cu, respectively.

3.3. Impact of street cleaning

Several approaches were used in order to identify any possible effect of street cleaning on ambient PM_{10} concentrations. When averaging the daily gravimetric PM_{10} concentrations during days with and without street cleaning (Table 1), no decrease was found. Conversely 12 and 17 μg/m^3 increments were found respectively at VAL1 and VAL2 during StC days, due to the higher regional pollution during the days in which StC was performed. However, if any benefit

Table 3
Literature comparison for the emission factors (μg veh^{-1} km^{-1}) of brake-wear related elements.

	Spain	Sweden	Switzerland	Austria	Austria	Sweden	Sweden	Lithuania	California
In PM10 (in PM2.5)	This study	Johansson et al. (2009)	Bukowiecki et al. (2009)	Laschober et al. (2004)	Handler et al. (2008)	Kristensson et al. (2004)	Sternbeck et al. (2002)	Valiulis et al. (2002)	Gillies et al. (2001)
Fe	7400 (4884)		4893		3400 (770)	(697)			12,390 (2790)
Cu	486 (306)	542	331	30	156 (41)	(0.2)	147–172	159 (98)	530 (170)
Sb	86	144	51		100 (8)		32–51		220 (150)
Sn	106	126	51		25 (7)				70 (100)

would take place, this should be still detectable when comparing with the COR reference station, but here the increment was only 9 μg/m^3 (Table 1) revealing that, on a daily basis, a worsening of PM_{10} concentrations took place at the cleaning site.

A further comparison was carried out with the local air quality monitoring network. The daily increments registered at VAL1 and VAL2 were respectively within 12–14 and 16–19 μg/m^3 and, in most cases, smaller than those registered at the reference site. Nevertheless no definitive conclusions can be drawn by these mean values. Within the EFs calculation, it was observed that for each pollutant (PM, Fe, Cu, Sb and Sn) Δx_i were not significantly lower during StC days, but stayed within the interquartile range, revealing that, on a daily basis, street cleaning did not have a significant decrease of x_i concentrations. These results, according to previous studies (Ang et al., 2008; Amato et al., 2009b; ARPA, 2003; Dobroff, 1999), reveal that to evaluate the effectiveness of street cleaning on a single open-air pilot road, little information is provided by 24 h average observations. Indeed one cannot expect high benefits: even measuring in a street canyon (where dispersion is assumed to be low), ambient air is continuously mixed with the rest of the neighborhood and abating a part of emissions on a single road does not imply automatically a reduction of PM (mostly on a daily mean). As an example, if we assume that, conservatively, the whole emissions from resuspension were abated during our test, this would provoke a daily emission benefit of 2280 gPM_{10}, which is equivalent to exhaust emissions derived from 31,000 km travelled by the average local fleet. This represent only 0.1% of the total km travelled (in average) in one day in the city of Barcelona. Thus, assuming that ambient air is continuously mixed, this reduces significantly the opportunity to detect significant PM decrease, by only cleaning one single road.

For the above reasons, we investigated better time-resolved measurements of PM and specific tracer of road dust emissions. The inter-comparison between continuous PM_{10} optical measurements and gravimetric data showed a very good agreement ($r^2 = 0.83$; 0.93; 0.97 for COR, VAL1 and VAL2 respectively). Looking at the average daily evolution of PM_{10} levels at the reference sites (Fig. 4), it can be seen how the increase during StC days is mostly observed during rush morning hours (07:00–09:00) and also during night. This was likely linked to an increased atmospheric stagnation and/or higher road traffic emissions (e.g. more congestions, Fig. 5), therefore we decided

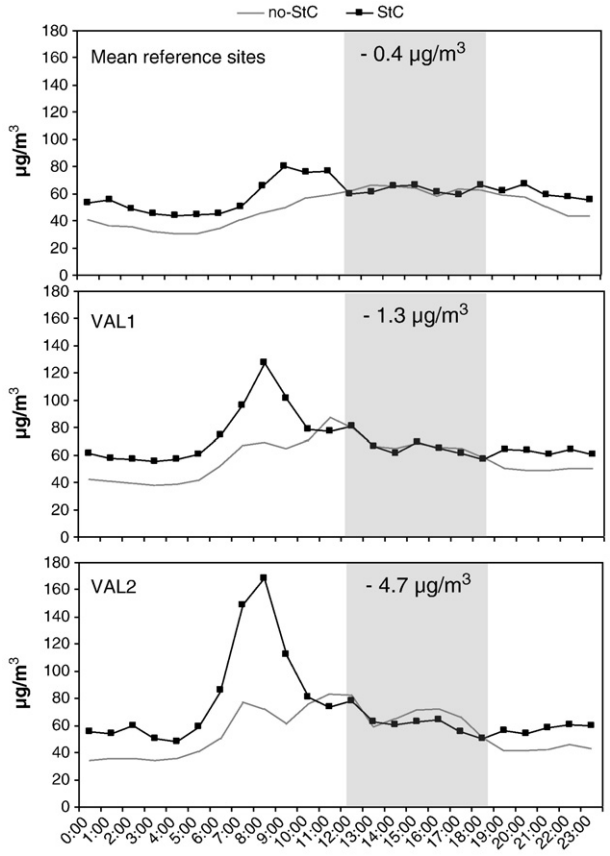

Fig. 4. Average daily evolution of corrected hourly PM_{10} concentrations at the two monitoring sites in Valencia Av. and the mean of reference sites, during days with street cleaning and without.

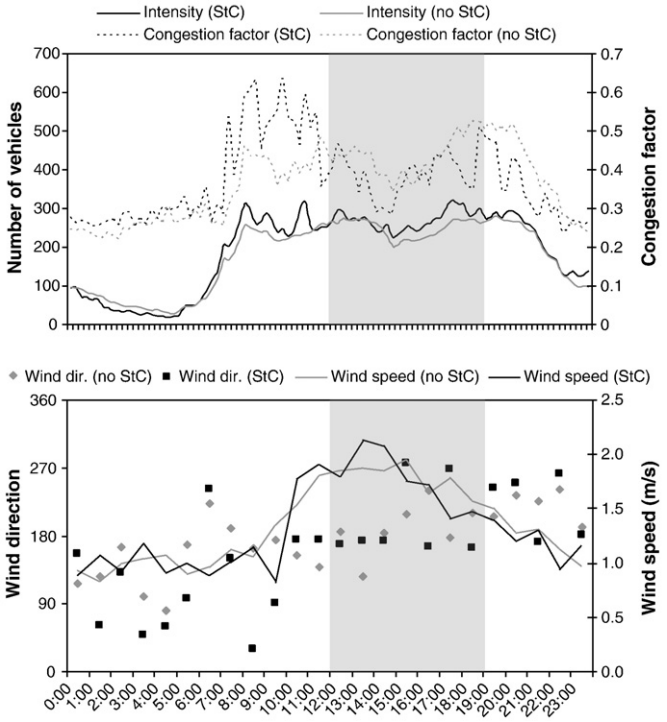

Fig. 5. Average daily evolution of traffic intensity and congestion [occupancy (‰)/intensity(#)] (top) and wind speed and direction (bottom) during StC and no-StC days.

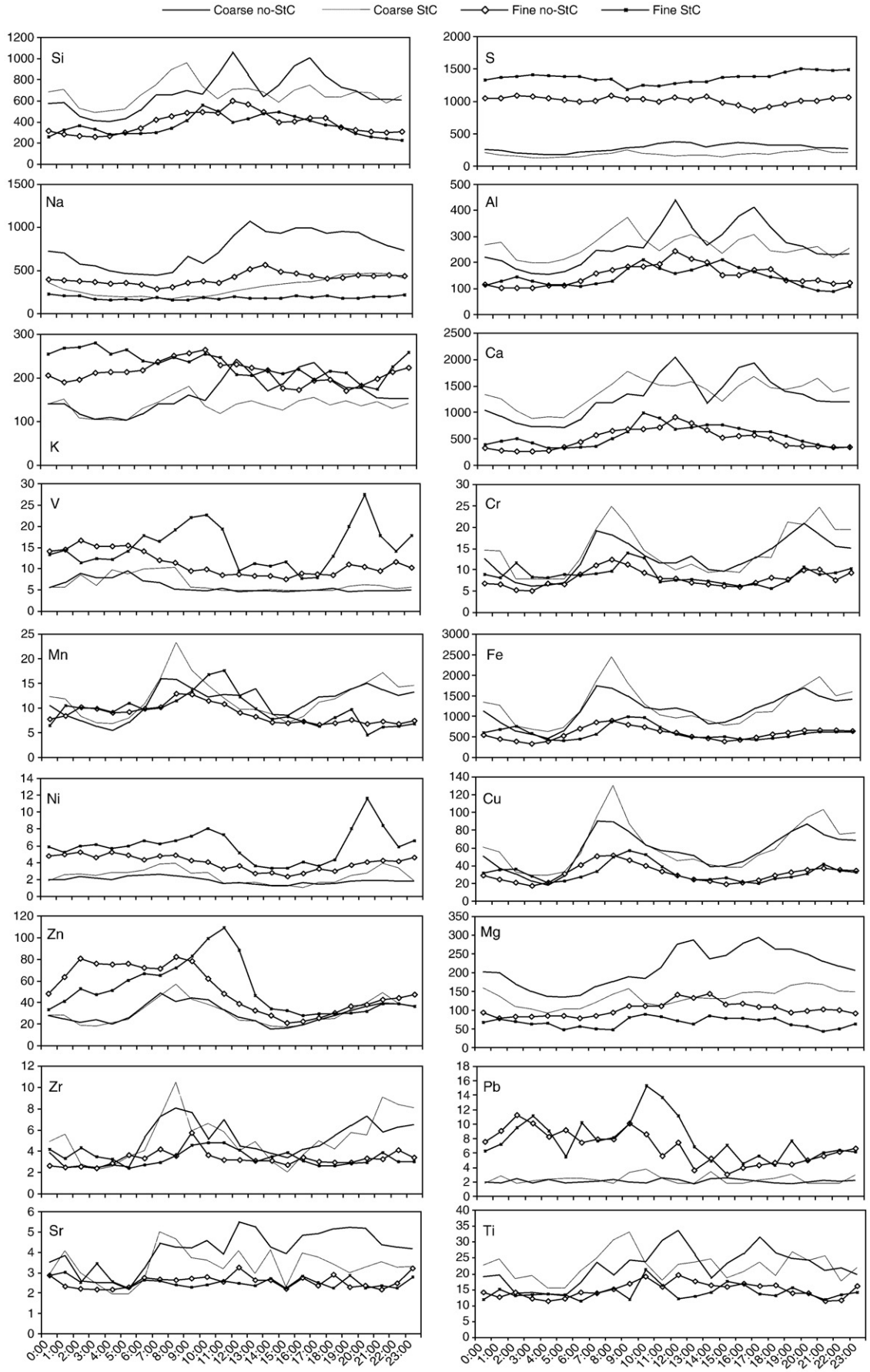

Fig. 6. Comparison of average daily evolution size segregated hourly elemental concentrations during StC and no-StC days.

to compare only the time frame between 12:00 and 18:00 which apparently was less affected by the worsening of urban and regional pollution (i.e. at the reference COR site no difference was observed between StC and no-StC days). During such hours, decreases of 1 and 5 μg/m^3 are observed respectively at VAL1 and VAL2 during StC days, in agreement with the downwind position of VAL2 with respect to the cleaned stretch. Given that the WSW sea-breeze transport was dominant, a smaller benefit at VAL1 was expected. However if a benefit occurred in afternoon hours, it should also took place in morning hours. The reason why it was not detected remains unclear. The increased traffic emissions during StC days might had some impact, provoking a worsening of PM levels more at VAL sites (50% more trafficked) than at the COR site.

The impacts of StC and meteorology on specific tracers were investigated by means of the hourly elemental composition of the $PM_{2.5}$ and $PM_{2.5-10}$ fractions. Unfortunately these data were available only for VAL1, which was less affected by the StC benefit during afternoon hours. During 3 weeks (before, during and after StC) hourly concentration of about 20 species were monitored, revealing different daily pattern for different type of PM_{10} components. Quality assurance of PIXE concentrations was assessed by comparing mean 24 h concentrations with those obtained by the chemical characterization of PM_{10} filter and calculating the r Pearson coefficient between them. Most of the species analyzed were in good agreement with ICP-MS, ICP-AES and HPLC results with the exception of Rb and Mo which were therefore excluded from the source apportionment study.

Sulfur showed a predominant fine particle size (82%). The coarse and fine fractions showed very different daily patterns (Fig. 6), being the fine S controlled by ammonium sulfate variability (regional processes) and therefore a reference parameter to investigate changes of regional PM pollution on a daily scale. The coarse fraction can be attributed to primary marine and secondary sulfate from the harbor activities showing a clear increase from 12:00 due to the channeling of the sea-breeze transport along *Valencia Av.* (similarly to Na and Cl). As shown in Fig. 6, by coincidence, in StC days atmospheric conditions were favorable for the accumulation of regional pollution (+34%) and probably also for the locally emitted pollutants. This is in agreement with the already mentioned PM_{10} increase at all reference stations during StC days (Table 1).

Iron, Cu, Zr and Cr showed a very similar daily evolution, with two peaks constantly at 9:00 and 21:00 due to fact that although traffic is fairly constant during the day and starts decreasing at 20:00, traffic related particles are cleansed by sea breeze from 11:00 to 20:00 h and register a peak only when wind speed depletes (21:00). The same pattern is observed for the two size fractions. During StC days, the two peaks were higher for all the elements revealing that both meteo conditions and road traffic emissions yielded to a more polluted scenario during StC days. Despite this, still a reduction is visible between 12:00 and 18:00 decreasing around 10% with respect to the same time frame in no-StC days. The fine fraction instead did not register any decrease.

Vanadium and Ni presented a very similar pattern for the fine fraction. Strong correlations between these two elements are widely documented in literature (Viana et al., 2009; Kim and Hopke, 2008) and are generally related to fuel oil combustion. Interestingly the fine fraction of Ni seemed instead more related to road traffic emissions.

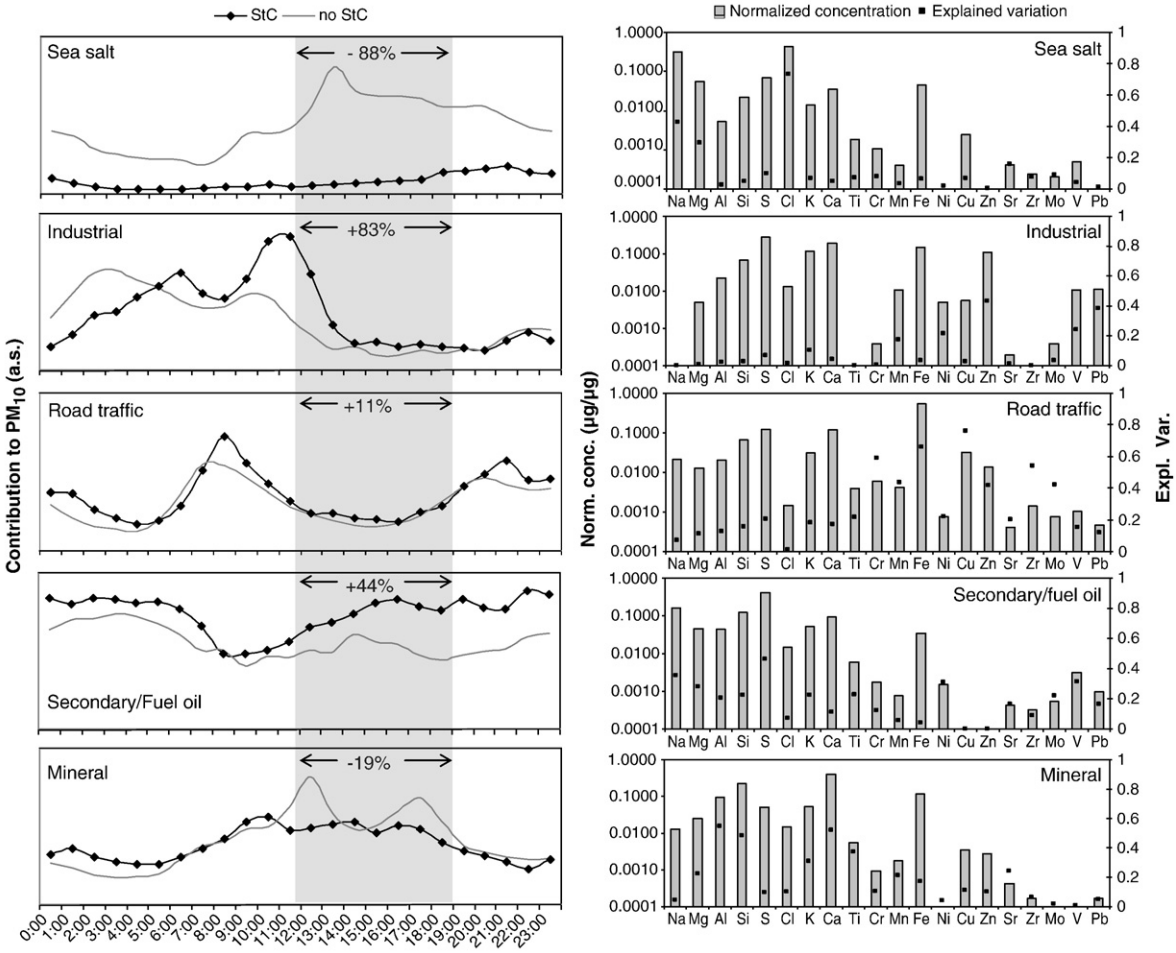

Fig. 7. Left: average daily evolution of PMF resolved source contributions (arbitrary scale) during StC days and no-StC days. Right: PMF source profiles.

As shown in Fig. 6 typical crustal elements, such as Si, Al, Ca, Ti and Sr, behaved in a similar way: typically higher concentrations during daily hours and for the coarse fraction two distinct peaks at 12:00 and 17:00 local time. Coarse particles were prevailing with respect to the fine (ratio coarse/PM_{10} within 0.56–0.72). During StC days a clear decrease for all these elements was observed varying from 12% (Ca), 18% (Ti), 22% (Al and Si) to 28% (Sr) but only between 12:00–18:00. Such decrease was less significant in the fine fraction (7–16%). A new peak also appeared at 9:00 probably related with works activity. Manganese and Mg behaved differently for the fine and coarse fraction, revealing two different main sources: a main mineral origin for fine fraction (similar to Al and Si) but a daily evolution for the coarse fraction driven by road traffic (Schauer et al., 2006; Amato et al., 2009c) and sea salt emissions respectively.

Lead and Zn showed high relative concentrations (respectively 61 and 73%) and daily pattern for their fine fraction, with maxima during night and minima during sea-breeze transport, revealing main possible source regions at the North. Coarse Zn showed a typical road traffic related pattern, similarly to Mn and Ni. No reductions could be associated with StC activities.

Fine and coarse concentrations for each element were then summed and submitted to a PMF source apportionment study with the aim of identifying the principal source of PM_{10} and their hourly evolution. Rotational ambiguity and validness of the proposed solution were explored by analyzing different FPEAK rotations (0.0 resulted the most realistic solution), several seeds, distribution of scaled residuals, G space plots and Q/Q_{exp} ratio (1.42).

Five factors were identified as primary road traffic PM, secondary aerosols, mineral, sea salt and industrial processes (Fig. 7). The road traffic factor represents primary exhaust and ferrous particles emitted from the vehicle wear (brakes, tires, disc etc). It explains more the 50% of the total variation of Cu, Fe, Cr and Zr. This factor also includes important concentrations of Zn, Mo and Mn (Fig. 7). The mineral factor, mainly carbonate and aluminum-silicate bulk, accounts for several sources of mineral matter, principally construction dust and traffic resuspension. The third factor, traced by S, Na, Ni and V, comprises those particles originated by the formation of secondary sulfate in atmosphere enhanced by atmospheric stagnation and photochemical oxidation of S (and presumably N) gaseous oxides. It also accounts for direct particulate emissions from fuel oil combustion, given the presence of the unequivocal tracers such as V and Ni. The industrial factor, with high concentrations of Pb and Zn was related to the influence of industrial activities located in the area, principally smelters. The sea salt factor is characterized by Na, Cl but also S and Mg. The sea salt and the industrial factor showed anti correlated daily patterns, due to the fact that industrial related emissions come predominantly from the northern industrial clusters.

As a coincidence a clear difference is noted between days with StC and without StC concerning contributions of sea salt and industrial particles. This is probably due to the increased sea-breeze transport during days without StC (see also Table 2). The secondary/fuel oil factor did register also an increase in contributions (44%) during StC days in the time frame 12:00–18:00.

Concerning the impact of road traffic emissions during the same period, an increase of 11% was also measured. Conversely, the mineral contributions were the only one registering a decrease (−19%) in StC with respect to days without StC. Therefore, during the StC days a slight decrease in mineral contribution at VAL1 was observed between 12:00 and 18.00 despite a general increase in PM_{10} levels due to enhanced atmospheric stagnation and traffic emissions. The overall decrease in the fitted contributions can be estimated at 2 $\mu g/m^3$ (3% of PM_{10} levels) but only if the 12:00–18:00 time frame is averaged. This result appears of low significance when weighted with the uncertainty of measurements, but it is interesting to note the agreement with the results from the continuous PM_{10} monitoring. Furthermore, according to the continuous PM_{10} measurement, this slight decrease is supposed to increase to 4–5 $\mu g/m^3$ at the downwind site.

4. Conclusions

The results evidenced the key role of meteorology in controlling PM variability and the low suitability of daily averages for evaluating the effectiveness of street cleaning. Meteorology resulted to have an impact on ambient air pollutant concentrations as strong as the one of source emissions. During the street cleaning week, atmospheric conditions favored stagnation and build up of pollutants, yielding to 20% and 34% mean increases of PM_{10} and NOx concentrations (respectively) at the reference street canyon site. Similarly, at the road where intensive street cleaning trials were undertaken (*Valencia Av.*), a mean daily increase of 15 $\mu g\ m^{-3}$ of PM_{10} was registered and comparison of daily averages with the reference sites did not offer evidence of a PM_{10} benefit at *Valencia Av*.

On the contrary, higher time-resolved concentrations, both for PM_{10} and trace elements mass, revealed that, during days of intense street cleaning, from 12:00 to 18:00, PM_{10} decreased at *Valencia Av* of 1.3 and 4.7 $\mu g\ m^{-3}$, at the upwind and downwind sites, respectively, whilst levels did not decrease at the reference sites. These values have high importance taking into account that the maximum emission benefit from street cleaning was calculated to be equivalent only to <1% of the total exhaust daily emissions in the whole city.

Moreover, the hourly elemental characterization of fine and coarse PM permitted to associate the small PM_{10} decrease during afternoon to mineral coarse particles. PIXE measurements showed in fact a clear decrease from 12:00 to 18:00 of coarse Si, Al, Ca, Ti and Sr within 12–28%, and less significant for the fine fraction (7–16%). Brake-wear elements (Fe, Cu, Cr and Zr) increased, on a daily average, during StC days, but from 12:00 to 18:00 showed also a decrease (around 10%) but only in the coarse fraction.

These results showed that, despite the meteorological noise and the low amount of abated emissions, there is some evidence of an air quality benefit induced by street cleaning activities. This finding is of great interest for air quality managers, given that any measure taken by local authorities for air quality improvement should be demonstrated and its benefit quantified when reporting to European Commission (Umweltbundesamt, 2006). However more suitable results would come from cleaning test on larger area (1–2 km^2).

Real-world Emission Factors were estimated for PM_{10}, Fe, Cu, Sn and Sb by the finding of a linear relationship between their increments and the increases of traffic emissions. The PM_{10} resuspension EF, averaged for the local fleet resulted to be 0.097 g $veh^{-1}\ km^{-1}$, and 1.6 times the exhaust EF, confirming the air quality burden of non-exhaust emissions in the Mediterranean region. For brake-related elements, total EFs were estimated, accounting for the sum of direct and resuspension emissions, in 7400, 486, 106 and 86 $\mu g\ veh^{-1}\ km^{-1}$, respectively for Fe, Cu, Sn and Sb. In $PM_{2.5}$Fe and Cu emission factors were respectively 4884 and 306 $\mu g\ veh^{-1}\ km^{-1}$. These findings are, to our knowledge, the first estimates of Emission Factors for non-exhaust emissions in the Mediterranean region and necessary tools for an exhaustive management of PM sources in Mediterranean cities.

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