Effects of water and CMA in mitigating industrial road dust resuspension

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Keywords: PM10, road dust, soil, mineral, crustal, CMA

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ABSTRACT

Water spraying and/or chemical suppressants such as salts and polymers have been suggested to reduce road dust resuspension due to their capability to increase adhesion, and therefore the effective size and weight of particles, but contrasting results have been obtained so further testing are needed. This study presents the first results of street washing and Calcium Magnesium Acetate (CMA) efficiencies at two industrial roads (paved and unpaved) in the Mediterranean region where the high solar radiation, warm climate, and scarce precipitation, may play a key role in determining the efficiency of mitigation techniques. Results show that, at

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both sites, street washing (water only) was more effective than CMA. Street washing made observe 18% (daily basis) and >90% (first hour) reductions of kerbside PM10 concentrations for the paved and unpaved road respectively, while with CMA PM10 decrease was generally lower and with less statistical significance.

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1. INTRODUCTION

Mineral dust (MD) is one of the main sources of atmospheric particulate matter (PM) at a global scale (Zender et al., 2004) dominated by desert dust contributions, and typically contributes 4-20% to the mass of ambient air PM_{10} in Central and Northern Europe and 4-34% in Southern Europe, with an increasing impact from rural to traffic locations (Putaud et al., 2004, 2010; Querol et al., 2004; Hueglin et al., 2005; Yin and Harrison, 2008) due to anthropogenic emissions, more important at local scale. MD concentrations depend on the geographical region and the local characteristics (geology, soils, land use, and meteorology) but also on site type and anthropogenic activities (road traffic, soil vegetal cover, urban works and industries). At specific receptors, local features may cause high soil dust particles concentrations causing severe individuals exposure and exceedances of PM air quality standards. Fugitive emissions, such as those arising from large construction works, cement and ceramic manufacturing, mining, and road transport, are very often left out of emission monitoring and inspections in Europe (Santacatalina et al., 2010). Consequently many urban agglomerations suffer from considerable impact of fugitive PM emissions, whereas significant improvements have been made in the implementation of Best Available Techniques for channeled emissions (Minguillón et al., 2007, 2009, 2013; Cuccia et al., 2011; Escudero et al., 2012; Moreno et al., 2014; Amato et al., 2014a). Spraying water and/or chemical suppressants has been suggested as mitigation measure due to their capability to increase adhesion, and therefore the effective size and weight of particles, but the effectiveness of these measures is not definitively demonstrated (Amato et al., 2014b; Norman and Johansson, 2006; Edvardsson et al., 2012). Calcium-Magnesium Acetate (CMA) has been tested as dust binder on paved roads in Sweden (Norman and Johansson, 2006), Austria (www.life-cma.at), Germany (Reuter, 2010), UK (Barratt et al., 2013) and Spain (Amato et al., 2014b). Other studies are available in Sweden, Finland and Norway (Gustafsson et al., 2010, 2013). Encouraging results were obtained in Sweden and Austria (-35% on daily mean PM10 concentrations), where road dust emissions represent a major contributor to PM10 levels, whilst studies in Germany, Spain and UK could not detect a significant PM10 decrease in typical urban roads, likely due to the higher degree of road moisture (Amato et al., 2014b). This is a crucial aspect, mostly in South Europe.

Two previous publications (Amato et al., 2009, 2014b) presenting the results of street washing, CMA and $MgCl_2$ application at a typical urban road in Barcelona (Spain), showed that street washing reduced on average PM10 levels by 7-10% (daily mean). CMA and $MgCl_2$ were not found to be effective in contrast to results obtained in Scandinavian and Alpine regions. This discrepancy suggests that effectiveness of dust suppressants is strongly influenced by dustiness of the road, type of pavement, solar radiation and road moisture. For this reason it is crucial to evaluate the effectiveness in more severe scenarios in Southern Europe, with higher road dust loadings and emission strength (Gustafsson et al., 2012). This study presents the first results of street washing and CMA efficiencies at two industrial roads (paved and unpaved) in order to discriminate whether road dust loading is a critical parameter to determine the effectiveness of CMA in the Mediterranean region.

2. METHODS

In this study, two different campaigns were carried out:

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- Campaign A: Evaluating the effectiveness of street washing and CMA at an industrial-urban paved road. The paved road is located within the ceramic industrial site of L'Alcora (Eastern Spain) and measurements were performed within June-July 2014. Effectiveness was calculated as the difference (in %) of PM10 ratios (test/control sites) between control and test days.

- Campaign B: Evaluating the effectiveness of street washing and CMA at an industrial unpaved road. The unpaved road was located within a quarry and concrete plant in Castellón (Spain), used for transportation of quarry materials by means of dumpers (July 2014). Effectiveness was calculated as the difference of PM10 concentrations between control and test days.

Figure 1. Map of North-Eastern coast of Spain with location of the two campaigns locations: L'Alcora (urban-industrial paved road, Campaign A), Castellón (industrial unpaved road, Campaign B)

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2.1 Urban-Industrial paved road (Campaign A)

The ceramic cluster of Castellón (NE, Spain) gathers 40% of EU, and 17% of worlwide ceramic production; PM10 levels have often exceeded European standards (Minguillón, 2007 and 2013) due mostly to MD (5–12 µg m^{-3} at urban sites). Most of the companies (250) are situated in 200 km^{2} forming a so-called *industrial cluster* that consumes 12 Mt year^{-1} of clay and 0.9 Mt year^{-1} of ceramic frits, glazes and pigments (Escrig et al., 2009). Raw materials suppliers for these companies are also located in this area. There are two opencast clay quarries and around 20 companies producing frits and other glaze components within this ceramic production region.

Two monitoring sites were selected for this study in L'Alcora, a small village (11,100 inhabitants) located 20 km in the inland, at the Western part of the ceramic cluster, with annual 31–36 µg m^{-3} of PM10 (Escrig et al., 2009; Querol et al., 2007). The first station (UB) is a urban background site belonging to the air quality monitoring network, and the second one (HS) is a laboratory van installed in a narrow street canyon (Figure 1) that leads to a very dense industrial cluster which includes one of the largest factories producing spray-dried granule and tile. This road (HS) is composed by two lanes (one per way), and along 250 m is continuously delimited on the sides by buildings of industrial plants. The 24-hours samples (00-00 UGT) of PM10 were collected on quartz micro-fibre filters (Ø15cm, PALL) by means of a high volume sampler DIGITEL DH 80 (30 m^{3} h^{-1}). 70 PM10 sampling was carried continuously from 26th June to 31st of July 2014 and acid digested (5 ml HF, 2.5 ml HNO_3, 2.5 ml $HClO_4$) for the determination of major and trace elements by inductively coupled plasma atomic emission spectrometry (ICP-AES). A section of 1.5 cm^{2} was used for the determination of OC and EC by a thermal-optical transmission technique using a Sunset Laboratory OCEC Analyzer with a EUSAAR2 temperature protocol. NOx, O3, Equivalent Black Carbon and meteo data were also available. Due to the high road dust loadings (20-40 mg m^{-2} Escrig et al., 2011), much above

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the EU range in cities (Amato et al., 2011) street washing activities were intensive. From 07:00 to 12:30 am, firstly the operators “dig out” dust from road shoulders, then a road wet sweeper (METCAR) passed through all the carriage. Totally 40 m^3 (26 $l\ m^{-2}$) of phreatic water were used in each water cleaning day. During other, well-separated days, CMA was sprayed by means of a 12 $l\ min^{-1}$ sprayer at 7 am and operators lasted 1 hour to cover the 250 m long road stretch. Two different dosages of CMA solution were applied: 30 $g\ m^{-2}$ of CMA + 0.03 $l\ m^{-2}$ of cold water (one test), and 60 $g\ m^{-2}$ of CMA (four tests).

2.2. Industrial unpaved road (Campaign B)

The study was located within a quarry for concrete/cement production with a road exclusive for the transport of the blasted material to the grinder with dumpers (Figure 1). Three dumpers were used (3 h^{-1}). Watering was the usual mitigation measure by means of a special-purpose irrigation truck, taking about 45 min to irrigate anew each part of the road. It could be readily observed that this strategy was quite effective in reducing the emissions.

The experiments conducted in this study were aimed to evaluate the life time of watering effect, and whether the watering effect could be enhanced by the use of CMA. Therefore, first, a known amount of water (see below) was applied on a selected stretch, then the watering was interrupted in order to quantify the life-time of effectiveness (i.e. the time needed for the road dust emissions to become significantly increased, similar to those before treatment). This experiment was repeated two additional times in which watering was complemented with the application of CMA by two different application modes. The selected stretch was about 125 m long and 9 m wide. Four continuous dust monitors (GRIMM and TSI Dust track) were placed downwind the stretch. The horizontal components of wind velocity were measured with a sonic anemometer. The incoming and reflected shortwave radiation was measured with an albedometer; the heat flux conducted through the ground surface was determined with a fluxmeter; and ground temperature was measured with a thermocouple.

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In the watering experiment, approximately 4000 l (3.5 l m^{-2}) of water were applied to the selected stretch. This amount of water was selected because it appeared large enough to obtain a homogeneous application throughout the stretch. It was approximately twice the load routinely applied by the company.

The two subsequent tests included CMA: the second experiment consisted of integrating 120 kg of CMA (i.e. a dosage of approx. 100 g m^{-2}) into 4000 l of water in the tank of the truck, although possible overdosage of water have occurred; in the third experiment the surface was first moistened with water (as usual procedure in the quarry), and then, 120 kg of CMA was subsequently sprayed onto the road surface with the same equipment already described in Campaign A.

3. RESULTS AND DISCUSSION

3.1 Urban-Industrial paved road (Campaign A)

During the campaign, mean daily concentrations of PM10 ranged within 40-161 µg m^{-3} at the HS site with a clear weekday/weekend difference (Figure S1), and within 6-31 µg m^{-3} at the UB site. The intra-daily variation of PM10 concentrations at HS showed a clear morning peak at 7 am, in coincidence with the highest traffic intensity and lower wind speed. From 10 am, PM10 levels decreased progressively due to the activation of sea breeze.

The evaluation of street washing and CMA was performed comparing daily and hourly averages across four sub-sets of data: i) control dry working days, without any treatment, ii) days with water cleaning, iii) days after water cleaning, iv) days with CMA. In order to minimize the effect of meteorology, the HS/UB ratio of PM10 concentrations was used. For hourly data,

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since no optical particle counter was available at UB, hourly PM10 at HS were normalized by hourly NO_2, EBC and wind speed measured at HS (Figure 2). The HS/UB ratio during working days (4.6) was much higher than for Saturdays (2.7) and Sundays (2.4), the average trucks intensity varying from 267, 44 to 0 trucks day^{-1}, respectively. This reveals the high impact of road dust resuspension, which was also noticeable visually.

Daily PM10 HS/UB ratios show a mean decrease of 18.5% of PM10 concentrations during the day of the water cleaning activities with respect to control working days (Table 1). However, this reduction was short lived, being reduced only to 2.2% the day after. This is probably linked to the high deposition rate in the tested road, estimated to be higher than 40 mg m^{-2} h^{-1} due to the continuous spilling of dusty materials from trucks (Escrig et al., 2011). For days with CMA application, the normalized PM10 concentrations show a mean daily decrease of 8.3% of PM10 concentrations in the days with CMA spraying (Table 1), being the decrease of concentrations mostly at morning hours (see below). Therefore, CMA effectiveness was lower than that of street washing (water only).

In order to validate such conclusion, the same evaluation was repeated for the normalized MD concentrations, which at the receptor site can be assumed to be a surrogate of road dust resuspension (88% increase of MD at HS with respect to UB). MD was calculated as:

$$MD = Al*3.89 + Ca*2.5 + Fe*1.43 + K*1.21 + Ti*1.67$$

in order to account for oxides, carbonates (calcite) and silica ($3*Al_2O_3$). Results in Table 1 show a mean daily reduction of 36% for MD during street washing days. Results show a mean reduction of 24% during CMA days, which again correspond to lower effectiveness than street washing. Table 1 also shows changes in elements concentrations across the different periods. The two-sample Wilcoxon rank-sum (Wilcoxon, 1945; Mann and Whitney, 1947) test was performed to HS/UB ratios for PM10 and its components analyzing separately control vs. CMA and control vs. water groups. For water cleaning, pollutants decreases were statistically

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significant ($p<0.05$) for Al, Ca, Mn, Zn and the total MD (Table 1). For CMA, decreases were statistically significant ($p<0.05$, Table 1) only for Al, Ca and Fe.

The main PM10 decrease during street washing and CMA days occurred from 7 am to 12 am, which correspond to the 5 hours after the beginning of treatment (Figure 2). Albeit short-lived, the decrease due to water cleaning was sufficient to produce a significant PM10 reduction also in the daily average (18.5%, Table 1). In fact, averaging the 5 hours from 7 to 11 am, PM10/NO_2 ratio decreased from 3.6 to 1.7 for water cleaning and only to 2.9 for CMA. PM10/EBC ratio decreased from 143 to 105 for water cleaning and increased to 147 for CMA spraying, revealing important reductions in the first 5 hours, but always more important for water cleaning rather than CMA (Figure 2).

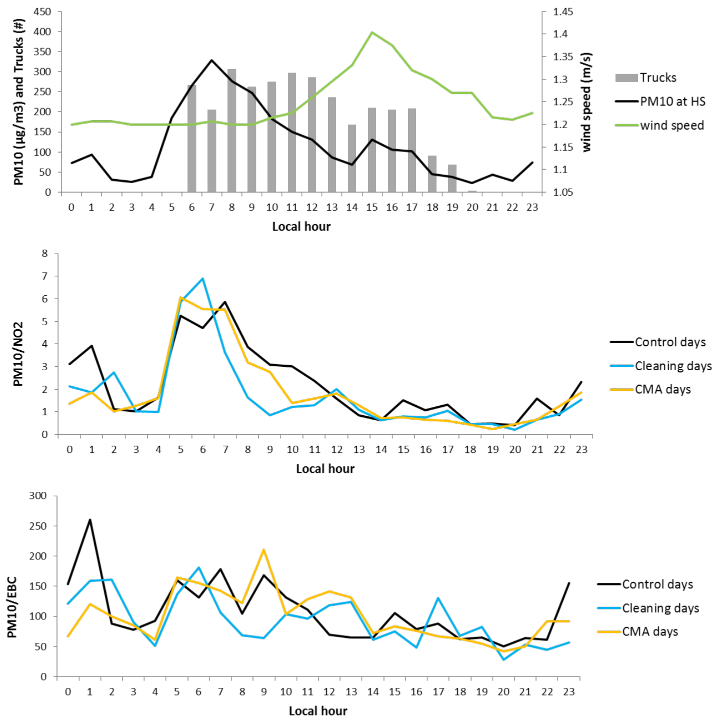

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Figure 2. Campaign A: Intra-daily variation of PM10, wind speed, traffic intensity, PM10/NO_2 and PM10/EBC at HS during street washing and CMA tests in the urban-industrial paved road.

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3.2. Industrial unpaved road (Campaign B)

Figure 3 depicts the recorded PM10 time series measured at the unpaved industrial road with dust monitors during the three different experiments, starting from the instant in which watering (left panel), watering+CMA (center panel) or CMA (right panel) application took place.

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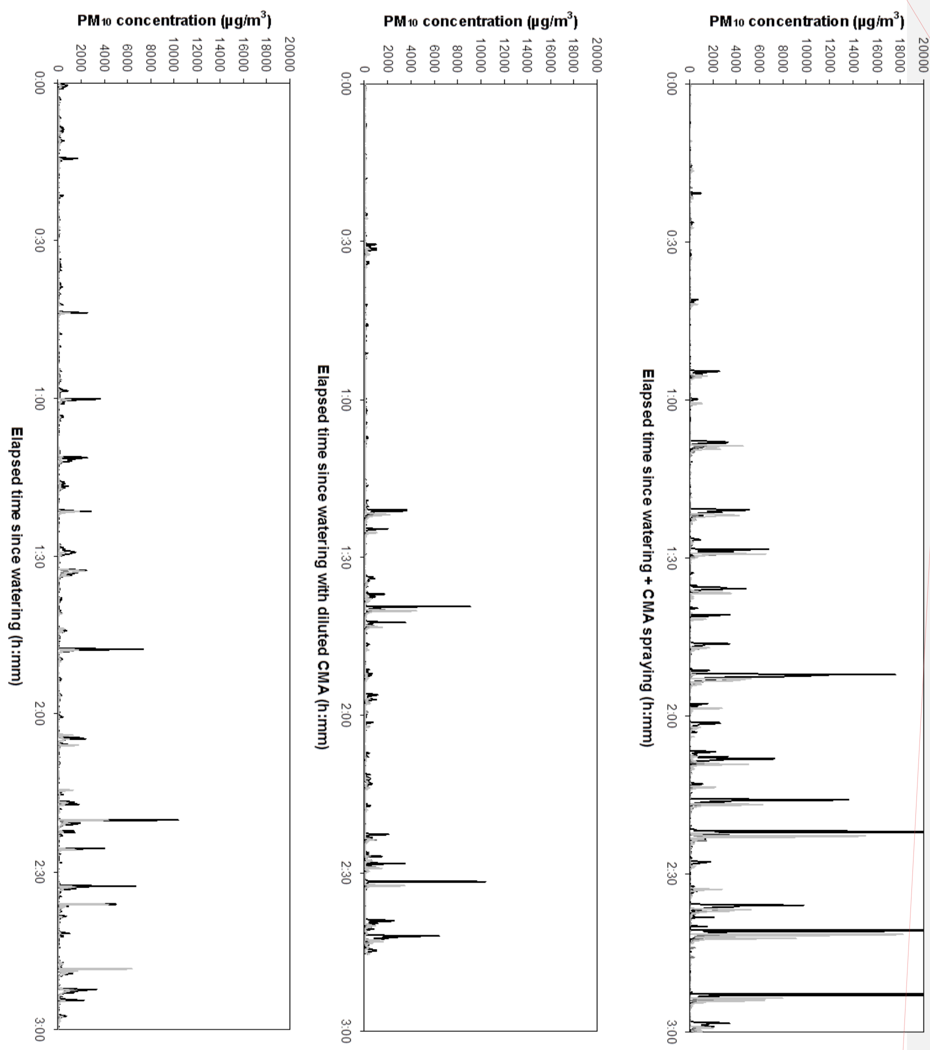

Figure 3. Campaign B: PM10 concentration time series (µg m^{-3}) obtained with two dust monitors (in black and grey) and snapshots of different events during the watering (left panel), watering+CMA (center panel) and CMA (right panel) experiments.

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In Figure S2 the time variation of the concentrations is complemented with additional relevant information. During the watering experiment wind direction was strictly dominated by the sea breeze throughout the experiment (130º) and wind speed remained quite constant at around 3 $m\ s^{-1}$. Incoming solar radiation ranged from 700 to 1000 $W\ m^{-2}$, and ground temperature from 40 to 45ºC.

The PM10 concentration time series obtained in the second (watering+CMA) experiment are shown in Figure 3 (center panel). Concentration peaks appeared to be slightly lower than those obtained in the watering experiment (left panel). In fact, when conducting the experiments it was perceptible that the road surface remained moistened longer than in the experiment of watering with water alone. However, this effect could not be readily attributed to the CMA, since, as indicated, the amount of applied water was larger. Furthermore, the experiment of watering+CMA was conducted in a cloudier day than the experiment with water alone. In sum, the effect of CMA, if any, was subtle. This might be attributed to the high amount of water used, which may have caused the run off of CMA, a third experiment was conducted in which (as reccomended by the CMA supplier) the surface was first moistened with the tucker truck, though in this case the amount of water was that applied usually by the company and then, 120 Kg CMA (100 $g\ m^{-2}$) was subsequently sprayed onto the road surface with the same equipment already described in Campaign A. This third experiment started earlier in the morning, and solar radiation, ground temperature, and wind speed were lower than in the other experiments (Figure 3, right panel). In spite of this, PM emissions became increased earlier than for the first experiment (watering) and the measured concentrations were higher. This outcome was attributed to the lower amount of applied water. The CMA spraying did not exhibit any apparent benefit (Figure 3, right panel).

The various tested measures exhibited all a high efficiency when applied, which was progressively reduced over time. In the cases where the larger amounts of water were

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involved (watering, and watering+CMA), its effect, though continously decreasing, appeared to remain even after the selected sampling period. CMA spraying was the less effective measure, while watering appeared to effectively reduce PM10 kerbside concentrations by more than 90% up to about 1 h after the application. Nevertheless, as already mentioned, this period is likely to depend on numerous factors; in particular, those that influence water evaporation rate.

Table 1. Campaign A: Average concentrations of PM10 components measureded at HS (tested paved road) and UB (control urban background site) sites during road dust mitigation tests. Statistically significant ($p<0.05$) reductions are indicated with *.

	HS				UB				HS/UB			
$\mu g\ m^{-3}$	Control	Weekend	Water cleaning days	CMA days	Control	Weekend	Water cleaning days	CMA days	Control	Weekend	Water cleaning days	CMA days
PM10	117.7	56.8	103.2	100.0	23.2	22.7	25.7	22.1	5.1	2.5	4.0	4.5
Al	7.71	3.17	6.80	6.71	0.52	0.56	0.72	0.61	14.9	5.7	9.4*	11.1*
Ba	0.05	0.02	0.05	0.05	0.01	0.01	0.01	0.01	9.3	4.2	7.6	8.8
Ca	6.26	2.40	5.97	5.26	0.76	0.71	1.21	0.92	8.3	3.4	4.9*	5.7*
Cu	0.0149	0.0057	0.0135	0.0152	0.0030	0.0026	0.0030	0.0030	4.9	2.2	4.6	5.0
Fe	3.06	1.26	2.87	2.58	0.26	0.27	0.34	0.28	11.9	4.7	8.5	9.3*
K	2.41	1.17	2.18	2.23	0.27	0.28	0.29	0.24	8.8	4.2	7.4	9.1
Mg	1.38	0.50	1.09	1.08	0.19	0.15	0.16	0.15	7.1	3.3	6.9	7.3
Mn	0.0297	0.0119	0.0267	0.0244	0.0035	0.0037	0.0048	0.0037	8.6	3.3	5.6*	6.5
Na	1.56	1.06	1.04	1.21	1.07	0.67	0.51	0.56	1.5	1.6	2.0	2.1
Ni	0.0049	0.0025	0.0026	0.0013	0.0021	0.0018	0.0016	0.0009	2.3	1.4	1.6	1.5
P	0.064	0.035	0.059	0.058	0.016	0.017	0.017	0.015	4.0	2.1	3.4	3.8
Pb	0.11	0.09	0.11	0.25	0.01	0.01	0.01	0.01	16.2	11.3	10.0	32.5

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S	2.16	1.86	2.15	1.80	1.17	1.00	1.20	0.78	1.8	1.9	1.8	2.3
Sr	0.022	0.009	0.020	0.019	0.003	0.003	0.004	0.003	7.8	3.4	5.5	6.9
Ti	0.35	0.14	0.31	0.28	0.03	0.03	0.03	0.03	13.2	5.5	9.1	9.8
V	0.012	0.007	0.011	0.008	0.005	0.004	0.004	0.002	2.5	1.7	2.5	3.6
Zn	0.15	0.11	0.16	0.15	0.02	0.03	0.04	0.02	7.3	3.7	4.5*	6.1
Total Mineral dust	81.70	33.34	73.51	70.63	6.53	6.72	9.36	7.61	12.5	5.0	7.9*	9.3
OC	5.45	3.47	4.80	3.95	2.38	2.36	2.19	1.90	2.3	1.5	2.2	2.1
EC	1.26	0.70	1.24	0.92	0.34	0.29	0.37	0.34	3.7	2.4	3.3	2.7

4. CONCLUSIONS

After testing street washing and dust suppressants on a typical urban paved road the AIRUSE LIFE+ project carried out further tests in order to evaluate water cleaning and CMA at paved and unpaved industrial roads in the province of Castellón (East of Spain) with a much higher contribution of local road dust PM10 emissions. Table 2 summarizes the combined results of this study with those obtained from previous studies (Amato et al., 2009 and 2014), in order to provide a quick overview of street washing and dust suppressant efficiencies in a typical Mediterranean climatic region.

Table 2. Summary of AIRUSE LIFE+ efficiency tests for road dust and soil dust mitigation

Source	Location	Dust loading	Measure	Dosage	PM10 reduction	Measurement site
Road dust	Urban paved road	3-6 mg m^{-2}	Street washing (Amato et al., 2009)	1 l m^{-2}	7-10% on a daily mean	kerbside
			CMA (Amato et al., 2015)	15-20 g m^{-2}	Negligible	kerbside
			$MgCl_2$ (Amato et al., 2015)	15-20 g m^{-2}	Negligible	kerbside

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	Industrial paved road (Campaign A)	20-40 mg m^{-2}	Street washing (present study)	27 l m^{-2}	18% on a daily mean	kerbside
			CMA (present study)	30-60 g m^{-2}	8% on a daily mean	kerbside
	Industrial unpaved road (Campaign B)	-	Street washing (present study)	3.5 l m^{-2}	>90% up to 1 h	downwind
			CMA (present study)	100 g m^{-2}	Not observed	downwind

Street washing was found to be the most effective measure in all the tested roads (urban paved, industrial paved and unpaved). Reduction on mean PM10 levels was estimated at 7-10% (daily mean), 18% (daily mean) and >90% (first hour) respectively, as measured at kerbside monitoring sites. For CMA and $MgCl_2$ efficiencies in PM10 were null or lower than that of water only (e.g. 8% for CMA versus 18% for water at the industrial paved road). The subtle effectiveness of CMA and $MgCl_2$ in Southern Europe (in contrast with Central and Northern Europe) is attributed to the high solar radiation, rapid evaporation of road moisture of the Mediterranean climate and consequently to the lower capacity of CMA and $MgCl_2$ to keep a high road moisture and bind road dust particles.

ACKNOWLEDGMENTS

This work was funded by AIRUSE LIFE+ ENV/ES/584 project. The study was also partially funded by Spanish Ministry of Environment through the Fundación Biodiversidad, with project

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Usuari 12/1/16 08:46
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Public park ... [4]

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Eliminado: In addition, a 10-20 nm polymer was tested over a 6200 m^2 unpaved area in a central urban park in Barcelona, where an air quality monitoring site has experienced several exceedances of air quality PM10 standards over the past years. In spite of the overall drop of PM10 concentrations across the city due to meteorological conditions and a general reduction of emissions, results allowed identifying an additional decrease of PM10 concentrations at the tested park site only, attributable to an inhibited resuspension of MD particles. The polymer-induced decrease, averaged over a 7 months period, was estimated at 2.9-4.5 µg/m^3 (up to 11% of PM10 concentrations). The effectiveness of the polymer in roads is however unknown.

acronym EMIDIF. The logistic support from the City Hall of Barcelona (Dept. of Investments and Transport Networks, Dept of Environmnetal Quality and Innovation), City Hall of L'Alcora, the Cantera La Torreta (grupo ORIGEN MATERIALES) company is also acknoweldgedFulvio Amato is beneficiary of the Juan de la Cierva postdoctoral grant (JCI-2012-13473).

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Eliminado: . The Barcelona Public Health Agency (ASPB) and the Government of Catalonia, for providing PM10 data and instrumental support.

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