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Discussions

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Trends of road dust emissions contributions on ambient PM levels at rural, urban and industrial sites in Southern Spain

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Abstract

The impact of road dust emissions on PM_{10} and $PM_{2.5}$ mass concentrations recorded from 2003 to 2010 at 11 locations (rural, urban and industrial) in Southern Spain was estimated based on the chemical characterization of PM and a the use of a constrained Positive Matrix Factorization, where the chemical profile of local road dust samples is used as a priori knowledge. Results indicate that road dust emissions increased PM_{10} levels on average by 21–35 % at traffic sites, 29–34 % at urban background sites, 17–22 % at urban-industrial sites and 9–22 % at rural sites. Road dust contributions to ambient PM levels show a marked seasonality with maxima in summer and minima in winter, likely due to the rainfall frequency. Decreasing concentrations trends over the sampling years where found at some traffic and urban sites but in most cases less significant than for vehicle exhaust emissions, while concentrations increased at industrial sites, probably due to local peculiarities. Concerning $PM_{2.5}$, road dust contributions were lower than in PM_{10} as expected, but still important (21–31 %, 11–31 %, 6–16 % and 7 % for traffic, urban background, urban-industrial and rural sites respectively). In addition the three main sources of road dust (carbonaceous particles, brake wear and road wear/mineral) were identified and their contributions to road dust mass loadings estimated, supporting air quality managers to drive measures aimed at preventing the build-up of road dust particles on roads.

1 Introduction

PM_{10} concentrations in large European cities over the last decade are not decreasing as expected (EEA, 2012; Harrison et al., 2008). This might be due to the underestimation (or absence) of important sources of primary PM in emission inventories (e.g. road dust) or to secondary aerosol precursors whose emissions reduction have not been substantial (e.g. NOx, VOC and NH_3). Kousoulidou et al., (2008) showed clear evidence that non-exhaust sources (road dust and wear emissions) become increas-

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ingly important as no emission control strategies are taken by member states. Road dust emissions are also pointed as the source responsible for the mismatch between modeled and observed PM_{10} concentrations in cities (Schaap et al., 2009 among others).

The most echoing impact of road dust emissions is the contribution to PM mass (parameter regulated by the EU Directive 2008/50/EC), due to their relatively coarser size distribution (typically between 1 and 10 µm), causing a high number of the exceedances of air quality limit values at urban and traffic sites. However, road dust is also of concern due to the high content of specific harmful components such as heavy metals and metalloids (i.e. Cu, Sb, Sn, Fe, Zn, Mo, Amato et al., 2009a), sulphides and carbonaceous aerosols such as elemental and organic carbon (EC and OC) and Polycyclic Aromatic Hydrocarbons (PAHs, Pengchai et al., 2004; Majumdar et al., 2012) among others. Heavy metals and sulphides originate from the erosion of brake and tire materials and induce oxidative stress (Yanosky et al., 2012). In California, a correlation between atmospheric concentrations of heavy metals (Fe, Cu, Zn, and Ni) and the mortality rate due to ischemic heart disease was recently found (Cahill et al., 2011). In Stockholm, Meister et al., (2011) estimated a 1.7 % increase in daily mortality per 10 $\mu g\,m^{-3}$ increase in $PM_{2.5-10}$ concentrations that include road dust and other coarse-size particles. The association with $PM_{2.5-10}$ was stronger for the November–May period when road dust was found to be most important. Exposure to an increase equivalent to the interquartile range of road dust contributions (below 2.5 microns) was associated with a 7 % increment of cardiovascular mortality in Barcelona (Ostro et al., 2011). Gustafsson et al., (2008) found that particles from road wear caused by studded tires are at least as inflammatory as particles from diesel exhaust.

Given these evidences, the actual pollution scenario is not encouraging. An overview of atmospheric concentrations of heavy metals in Spain (more than 20 monitoring sites) revealed that the highest concentrations of Fe, Cu, Sr, Sb, Ti and Ba (and partly also Zn and Zr) are measured inside the cities (rather than at industrial hotspots), where most of population live and work (Querol et al., 2007). Concentrations increase further at road-

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side locations where also a significant part of population is exposed. Consequently, for these metals, population exposure is much higher than for common industrial tracers such as As and Cd. Investigating the role of non-exhaust emissions in air quality impairment and their impact on health is therefore a non-regret policy and a must for local authorities, mostly considering that such particles are emitted locally, and are therefore easier to control/mitigate, improving public health.

The identification of individual source contributions is crucial for the understanding of health effects since low-contributing sources may be more relevant for health. Differentiating the contribution of road dust from other traffic sources is however problematic: the complexity of the urban environment does not always allow for a clear separation of road traffic sources, consequently most of source apportionment studies presented so far, show results only for total contributions from road traffic emissions (Viana et al., 2008). It is also common to find studies where the road dust component of traffic emissions is mixed with other mineral/soil sources. PM contributions from vehicular traffic should be differentiated between exhaust and non-exhaust fraction. Ideally non-exhaust contributions should be further separated between road dust, brake, tire and road wear.

The relative importance of these categories changes widely in space and in time. Spatially, road dust emissions increase largely in Southern Europe (due to drier climate) and Scandinavian countries (due to the road sanding and the use of studded tires) (Querol et al., 2004) but also within a city environment e.g. next to construction sites and in heavy traffic roads (Amato et al., 2009). Timely, road dust emissions are severely influenced by meteorology (precipitation, insulation, road humidity and droughts, Amato et al., 2012). In addition, it is important to monitor the relative increase of non-exhaust emissions (currently uncontrolled) against the motor exhaust emissions, which have been progressively reduced in last two decades by means of the EURO (1 to 5) standards.

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To date, research on non-exhaust emissions has been rather limited due to the difficulties encountered by experimentalists and modelers to characterize and describe the complex phenomenon of road dust resuspension and wear emissions.

In this study we aim to contribute to the improvement on current knowledge on road dust emissions by estimating their impact on PM_{10} and $PM_{2.5}$ levels measured at 11 receptors distributed at traffic, urban, industrial and rural location across Andalucía, the most arid and populated region of Spain, which suffers also of frequent Saharan dust deposition events.

2 Methods

2.1 Study area

Andalucía is the most populated region in Spain with 8 provinces and 8.4 million inhabitants. The local economy is basically based on tourism, being the primary (agriculture, fishing and mining) and industrial sectors only a small percentage of the gross value added. Urban road traffic is very dense due to the commonly insufficient public transport infrastructure (metro, tram, buses) and to the high density of urban architecture. Climate is typically Mediterranean, with dry and hot summers and mild winters, favoring the build-up and mobilization of road dust particles and their entrainment into the atmosphere due to the wheel and vehicle induced turbulence.

Due to the evident impact of road dust emissions and their increasing concern (Harrison et al., 2008; Denier van der Gon et al., 2013) on urban air quality the Regional Government of Andalucía had recently promoted research studies aimed at evaluating the impact of road dust emissions on air quality measured at large cities and industrial areas of Andalucía. The five cities under study (Seville, Malaga, Cordoba, Granada and the Algeciras Bay, Fig. 1) are briefly described below:

– Seville, the capital of Andalucía, is the fourth largest city of Spain, counting 700 000 inhabitants, and 1.5 million in the Metropolitan region. The city is built on

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the flat depression of Guadalquivir river and hosts the only inland port of Spain. Road traffic emissions have been partially reduced by the recent construction of tram and underground lines.

- Malaga is the second most populous city of Andalucía with a population of 600 000 (metropolitan area). It lies on the Mediterranean coast, 100 km east of the Gibraltar Strait, and it is bordered to the North by a high mountain range. Malaga is a city of commerce and tourism, therefore the local anthropogenic pollutant source is mainly traffic.

- Cordoba (330 000 inhabitants) is one of main touristic destination in Spain. The city is located on the banks of the Guadalquivir river and has the highest summer average daily temperature of Europe (36,2° in July). The climate is Mediterranean-Continental. Precipitations are concentrated in winter-autumn and summer is often characterized by droughts.

- Granada (738 m a.s.l.), is a non-industrialized and medium sized city with 250 000 inhabitants. The city is located in a natural valley surrounded by mountains with elevations between 1000 and 3350 m a.s.l. Traffic is the most important source of anthropogenic pollutants in Granada. The topography of Granada favors the development of thermal inversions in winter, with a significant accumulation of pollutants in the study area (Lyamani et al., 2008).

- The Algeciras Bay hosts the largest port of Spain (70 millions of tonnes), oil refinery and stainless steel plants. It is around 10 km long by 8 km wide. It opens to the south into the Strait of Gibraltar, where around 80 000 ships per year leave the Mediterranean into the Atlantic. Due to all these activities along the shoreline, air pollution is a severe problem in the area. Heavy trucks visibly dominate road transport. This area is also characterized by a high population density, with more than 300 000 inhabitants.

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2.2 Ambient air PM sampling

PM_{10} and $PM_{2.5}$ samples were collected at the five provinces under study from 2003 to 2010. Details of most of the monitoring stations considered in this work can be found in de la Rosa et al. (2010). Sampling periods and frequency varied among sites; details are shown in Table 1. Eleven stations were selected among rural, urban background, traffic and urban-industrial environments:

– In the Seville province, one traffic (Principes) and one urban background site (Alcalá de Guadaira), the latter being located in the suburb of the city were used (refs). Several studies on air quality have been already performed in metropolitan area of Seville (Adame et al., 2012; Notario et al., 2012).

– In the Malaga province, one traffic (Carranque) and one rural site (Campillo), located 60 km northwest of the city were chosen.

– In the province of Cordoba, one urban background (Lepanto) and one rural site (Poblado) were selected. (García Lorenzo, 2011). Details of the monitoring stations can be found in Lozano et al. (2009).

– In the city of Granada, the traffic monitoring site Granada Norte was used, located in between the two carriages of the Davalos avenue, counting 15 000 vehicles per day totally.

– In the Algeciras Bay four urban-industrial monitoring sites were used (Los Barrios, La Línea, Algeciras and Puente Mayorga). Details can be found in Pandolfi et al. (2011) and Gonzalez Castanedo (2011).

Daily PM samples were collected by means of high volume ANDERSEN (68 $m^3 h^{-1}$ onto rectangular filters) and MCV (30 $m^3 h^{-1}$ onto circular filters) samplers with a frequency of 1–2 samples per week at all sites. A total of 2696 filters were collected on quartz fiber filters (Schleicher & Schuell, Munktell and Pallflex). Before sampling, quartz

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fiber filters were dried at 205 °C during 4 h and conditioned for 48 h at 20 °C and 50 % of relative humidity. Weights of blank filters were measured three times every 24 h (or five times in case of rectangular filters) by means of a Sartorius LA 130 S-F microbalance (1 µg sensitivity). After weighing filters were kept in aluminum foils and brought back to laboratory to be weighted two more times every 24 h. Once the weights of samples were determined, filters were destined to several analytical (destructive) treatments. These procedure are briefly listed below, more details are available in Querol et al. (2001):

- Half filter (or 150 cm^2 in case of rectangular filters) was acid digested (5 mL HF, 2.5 mL HNO_3, 2.5 mL $HClO_4$) for the determination of major and trace elements and analyzed respectively by inductively coupled plasma mass spectrometry and atomic emission spectrometry (ICP-MS and ICP-AES) (Querol et al., 2001).

- A quarter of filter (or 75 cm^2 in case of rectangular filters) was leached in 20 mL of bi-distilled water for the extraction of water-soluble ions and subsequent analysis by ion chromatography (IC) for sulfate, nitrate and chloride and by specific electrode for ammonium.

- A section of 1.5 cm^2 of the filter was used for the determination of TC by means of elemental analysis.

In every case blank concentrations were subtracted for determining final concentrations in samples.

2.3 Road dust samplings

Road dust samples were collected, in each city, at four selected roads in the vicinities of the urban monitoring site used for ambient air PM. Road dust particles were collected on 47 mm diameter quartz fiber filters (Pallflex) by means of a dry vacuum sampler coupled with a 10 µm-inlet able to separate only the mobile particles with aerodynamic

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diameter < 10 µm from a delimitated area of active road surface. Details of the device and sampling protocol can be found in our previous publication where the mass loadings of road dust were investigated (Amato et al. 2013). Details of sampling sites are summarized in Table S1. At each road, three different samples were collected in order to improve representativeness and to collect enough sample for a complete chemical characterization:

- The first filter was acid digested (following the above protocol) to determine the concentration of major (Na, Mg, Al, Fe, P, S, Cl, K, Ca, Ti and Mn) and trace elements (Li, Sc, V, Cr, Co, Ni, Cu, Zn, Ga, Ge, As, Se, Rb, Sr, Zr, Nb, Mo, Cd, Sn, Sb, Cs, Ba, La, Ce, Hf, W, Tl, Pb, Bi, Th and U among others) by means of ICP-AES and ICP-MS respectively.
- One half of the second filter was used for a leachate in Milli-Q water (20 mL) for the extraction of soluble ions and subsequent Ion Cromatography analysis to determine the concentration of sulphate, nitrate, chloride and specific electrode for NH_4^+.
- A fraction of 1.5 cm^2 of the second filter was used for the determination of Organic carbon (OC) and Elemental carbon (EC) by means of the Sunset thermal-optical analysis (Birch and Cary, 1996).
- A fraction of 1.5 cm^2 of the second filter was used for the determination of particulate Hg by means of the atomic absorption spectrometer LECO AMA 254.

The third filter was stored for further analysis. For the source apportionment study, the average of the four chemical profiles obtained in each city was used.

2.4 Source apportionment

Positive Matrix Factorization (PMF, Paatero and Tapper, 1994) is a widely used model for atmospheric aerosol source apportionment, as well as for other types of samples

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such as ashes, soils etc. PMF is based on the mass conservation principle:

$$x_{ij} = \sum_{k=1}^{p} g_{ik} f_{jk} \quad i = 1,2,\ldots,m \quad j = 1,2,\ldots,n \tag{1}$$

where x_{ij} is the ith concentration of the species j, g_{ik} is the ith contribution of the source k and f_{jk} is the concentration of the species j in source k.

In this study two types of source apportionment analysis were carried out:

- a *road dust* source apportionment, in order to identify the main sources of road dust and their contribution to observed mass loadings. The *road dust* source apportionment, aimed at identifying the main sources of road dust was performed merging the 20 road dust samples obtained in this study with road dust samples from other Spanish cities (Barcelona and Girona, Amato et al., 2011) collected in previous studies from our group, in order to improve the statistical basis. Source contribution results are however presented only for the sites in Andalucía.

- a PM source apportionment, merging PM_{10} and $PM_{2.5}$ data for each area in order to estimate the contribution of main source of PM including road dust.

In both cases a PMF model was used. For the PM source apportionment a *constrained* PMF was carried out, divided in six different analyses due to the considerable distance among monitoring sites (i.e. the assumption of same sources at all sites may be not valid). The constraints consist of auxiliary equations (added to the main equations by means of the Multilinear Engine 2 programming, Paatero, 1999) in order to:

1. pull one factor profile f_{jk} towards the target profile of local road dust, obtained experimentally;

2. limit the daily g_{ik} $PM_{2.5}$ source contribution by the upper PM_{10} source contribution bound;

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3. limit the sum of factor profile by the maximum of 1.

This approach has been already followed by Amato et al. (2009b) and Amato and Hopke (2012) showing a satisfactory theoretical basis. The six analyses were divided as follows:

1. PM_{10} and $PM_{2.5}$ analysis at Los Barrios, Algecíras, La Línea and PM_{10} analysis at Puente Mayorga (four urban industrial sites of Algecíras Bay). A total of 1506 PM samples were used (298 PM_{10} and 157 $PM_{2.5}$ samples at Los Barrios, 343 PM_{10} and 289 $PM_{2.5}$ samples at la Línea, 79 PM_{10} and 82 $PM_{2.5}$ samples at Algecíras, 258 PM_{10} samples at Puente Mayorga) against 30 components of PM.

2. PM_{10} and $PM_{2.5}$ analysis at Poblado (rural) and Lepanto (urban background) stations (Cordoba). A total of 246 PM samples were used (175 PM_{10} and 50 $PM_{2.5}$ samples at Lepanto, 21 PM_{10} samples at Poblado) against 30 components of PM.

3. PM_{10} and $PM_{2.5}$ analysis at Príncipes (traffic) and Alcalá de Guadaria (urban background) stations (Seville). A total of 410 PM samples were used (170 PM_{10} and 44 $PM_{2.5}$ samples at Príncipes, 163 PM_{10} and 33 $PM_{2.5}$ samples at Alcalá de Guadaria) against 29 components of PM.

4. PM_{10} and $PM_{2.5}$ analysis at Granada Norte (traffic site in Granada). A total of 217 PM samples were used (183 PM_{10} and 34 $PM_{2.5}$ samples) against 31 components of PM.

5. PM_{10} and $PM_{2.5}$ analysis at Carranque (traffic) (Málaga). A total of 222 PM samples were used (175 PM_{10} and 47 $PM_{2.5}$ samples) against 29 components of PM.

6. PM_{10} and $PM_{2.5}$ analysis at Campillo (rural site in the province of Málaga). A total of 95 PM samples were used (54 PM_{10} and 41 $PM_{2.5}$ samples) against 26 components of PM.

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For each PMF analysis, the selection of species was based on the two-fold criterion of S/N ratio and % of data above detection limit (Amato et al., 2009b). Details are shown in Table S2.

Moreover, temporal trends of source contributions were analyzed at those monitoring sites where at least four years of data were available. The Theil–Sen method (Theil, 1950; Sen, 1968), available in the Openair software (Carslaw, 2012; Carslaw and Ropkins, 2012), was applied to the monthly averages to calculate the regression parameters of the trends including slope, uncertainty in the slope and the p value. The applied method yields accurate confidence intervals even with non-normal data and it is less sensitive to outliers and missing values (Hollander and Wolfe, 1999). Data were deseasonalized and all the regression parameters were estimated through bootstrap resampling. The slopes indicate how road dust contributions have changed through time and are expressed in units ($\mu g\,m^{-3}$) per year. The p values show whether the calculated trends are statistically significant. A statistically significant trend was assumed at the 90th percentile significance level ($p < 0.1$ or +), meaning that there was a 90 % chance that the slope was not due to random chance. p values > 0.1 indicate insignificant trends, whilst p values $= 0.01$ and 0.001 (or ** and ***) indicate high and very high significant trends, respectively.

3 Results and discussion

3.1 Road dust composition and sources

As mentioned previously, the chemical composition of road dust at each city was investigated with two main objectives:

– Identifying the main sources of road dust and estimate their contribution to road dust loadings ($mg\,m^{-2}$) measured on active traffic lanes (this section).

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– Estimating the contribution of road dust to the levels of ambient PM_{10} and $PM_{2.5}$ ($\mu g\,m^{-3}$) measured at the PM monitoring sites under study (Sect. 3.2).

Road dust loadings have been already discussed in Amato et al. (2013). Briefly, typical urban roads showed emission factors within 77–480 $mg\,veh^{-1}\,km^{-1}$ the averages 158 ± 90 $mg\,veh^{-1}\,km^{-1}$ in Córdoba, 180 ± 113 $mg\,veh^{-1}\,km^{-1}$ in Málaga, 189 ± 27 $mg\,veh^{-1}\,km^{-1}$ in Seville and 347 ± 144 $mg\,veh^{-1}\,km^{-1}$ in Granada. These values are in the upper edge of the range observed in Europe and US. An increasing trend of emission factors was observed from freeway, urban, urban-construction up to industrial sites. After averaging the elemental concentrations at the four roads under study at each city, results show that the main components of respirable (< 10 µm) road dust particles are OC, Ca, EC, Al_2O_3, Fe and Mg (silica was not analyzed) (Fig. 2). Concerning trace elements strong enrichments in Ti, Zn, Cu, Ba, Mn, Sn, Sb, Zr and Sr were found (Fig. 2). The relative proportion of these components may vary depending on the importance of the different sources involved: road wear, tire wear, brake wear, other minerals (unpaved areas, works) and motor exhaust. This results in a varying composition depending on the type of road (Table 2):

– Typically urban roads showed higher relative concentrations of OC, EC, Cu, Ba, Sn, Sb, Bi and W (usually produced by abrasion of brakes, road and tires) and Cl^-. Interestingly, a correlation was found between the loadings of Fe-Cu-Zn ($mg\,m^{-2}$) and the distance between sampling point and braking areas such as traffic lights and roundabouts (Fig.S1), regardless of roads category. This suggests that emissions of brake particles (both the airborne and deposited fraction) vary spatially within a city environment. This information is important for urban scale modeling and exposure studies.

– Industrial roads were enriched in Cr, Co, Ni, Pb, Mn and Cd (linked to PM deposition from stationary sources) and Ca (likely due to higher wear rate of road pavement by heavy duty vehicles).

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- Roads nearby works were enriched in Ca, Ti and Mg (due to the handling of constructing materials and hearths movements).

- Poor state of the pavement shows enrichment in Al, Ti, Rb and K, typical tracers of phyllosilicates.

The PMF identified three main sources responsible for the production and build-up of respirable road dust particles on road pavement: (i) a carbonaceous source, mainly related to tire wear, although a contribution of motor exhaust and brake wear may be present; (ii) road wear, which, in specific samples (Fig. 3) includes mineral dust from unpaved areas and works; (iii) brake wear. Factor profiles are shown in Figure S2. The sum of these three sources explains in average 96 % of the observed road dust mass loadings.

- The carbonaceous source is composed mainly by OC, and, in a minor proportion by EC and Ca, suggesting the presence of tire particles possibly attached to particles of calcite and bitumen from pavement. In average this is the main source of road dust particles (50 %). The highest contributions of this source were found in the Algeciras Bay area and in Granada. The typically urban contribution can be estimated in 3–4 $mg\,m^{-2}$ (Fig. 3).

- The road wear factor is responsible for the production of the mineral particles deposited on pavement, even though a contribution from unpaved areas and urban works cannot be discarded. The chemical profile is traced by typically crustal species such as Al, Ca, K, Ti, Fe and Mg, but this source is also responsible for a large variance of Pb and Hg. In typically urban roads, without nearby works, the contribution varies within 0.3–5.8 $mg\,m^{-2}$ with an average of 1.9 $mg\,m^{-2}$ (20 %, Fig. 3). In roads with nearby works, and/or unpaved areas, or with a poor state of the pavement, the contribution can reach up to 10 $mg\,m^{-2}$ (Fig. 3).

- The Brake wear factor is clearly identified for the high content in Fe (brake pads contain 13–45 % of metallic iron, Amato et al., 2012), Al and Ca. Aluminum is

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used as abrasive in pads (as metallic Al or corindon), while calcium is used in form of calcite as a filler. This factor also is responsible for most of the variance of Cu, Sn, Sb, Cr and Ba. All these elements are used in brake pads manufacture as lubricants (Sn and Sb sulphides), fillers (barite) and friction materials (metallic Cu, CuO and Cr_2O_3). Brake wear contributes in average 12 % of road dust particles, varying within 0.2–3.0 mg m^{-2} (Fig. 3). Compared to previous studies, road wear/mineral contribution are generally similar (in %) to other Spanish cities as Barcelona and Girona, while carbonaceous materials are higher as opposed to brake particles. Central European cities as Zurich and Utrecht showed much lower contributions from mineral matter (Amato et al., 2011 and 2013b).

3.2 PM source apportionment

For each PMF analysis the distribution of residuals, G-space plots, *F* peak values and *Q* values were explored for solutions with number of factors varying between 3 and 10. The most reliable solution identified six sources at all sites, with the exception of the Algeciras Bay area where eight sources were found (Fig. S3 and Table 3). Four sources were common at all sites, namely Road Dust (RD), Mineral (MI), Vehicle Exhaust (VE) and Secondary Sulfate (SS). Sea salt (SE) was found at all sites with the exception of the traffic station in Granada. Secondary nitrate (SN), Metallurgy (ME) and Heavy oil (HO) were identified at six of the eleven sites (Fig. S3 and Table 3). Tire wear (TW) contributions could be separated only at the traffic site of Granada Norte, where also a Traffic/Secondary (TS) factor, linked to atmospheric stagnation conditions, was found (Fig. S3 and Table 3).

The yearly average road dust contribution was found to increase from rural (9–22 % of PM_{10}), urban-industrial (17–22 %), urban (29–34 %) to traffic (21–35 %) PM_{10} levels (Fig. 4 and Table 3). Concerning $PM_{2.5}$, road dust contributions were lower but the same pattern is shown: 7 %, 6–16 %, 11–31 % and 21–31 % respectively (Fig. 5 and Table 3). However it has to be noticed that road dust contributions show a marked

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seasonality with maxima in summer and minima in winter, likely due to the rainfall frequency (Fig. 6).

The time trend analysis shows statistically significant decreasing trends for the road dust source at two traffic sites (in Granada and Seville, with *p* values of 0.5 and 0.01 respectively) and at the urban background site in Córdoba (p value 0.05) in most cases accompanied by stronger and more significant trends also for vehicle exhaust emissions (Fig. S4 and S5). Overall, the downward trend of road dust contribution at the traffic sites can be estimated in -1.5–$2.0\,\mu g\,m^{-3}\,yr^{-1}$ which can be related to the decrease in construction/demolition activities from 2008 on, due to the financial crisis. As road dust sampling showed, particles generated by urban works increase significantly the road dust mass loadings on road surface and consequently also road dust emissions. The downward trend is in fact also found for the mineral source found at Granada Norte, Príncipes, Lepanto, Carranque, Los Barrios and La Línea.

No trend was observed at the traffic site in Málaga and at the urban background site in Seville (p value > 0.1), although in Seville a clear decrease can be observed after 2009.

Increasing trends of road dust contributions were found at two of the three industrial sites (La Línea and Los Barrios) with very high statistical significance ($p=0.001$). At these two sites, motor exhaust and mineral contributions decrease significantly, while only road dust and (slightly) secondary nitrate showed increasing trends (Fig. S4 and S5). At the third site (Puente Mayorga) also a positive trend is shown but without statistically significance ($p > 0.1$). At the industrial sites, the increasing trend of road dust contributions is confirmed by the increase in Cu concentrations (a typical tracer for brake wear) by a rate of $4\,ng\,m^{-3}\,yr^{-1}$ ($p = 0.001$) and possibly due to local peculiarities such as the construction of new parking lot and urbanization in the vicinity of the monitoring site and the proximity to the national border between Spain and Gibraltar.

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4 Conclusions

In this study we estimated the daily contribution of road dust emissions to the PM_{10} and $PM_{2.5}$ mass concentrations measured across 2003–2010 at eleven air quality monitoring station in Andalucía through a constrained PMF analysis. The chemical profiles of local road dust samples were used as targets within the multivariable regression. Road dust emissions were found to contribute largely to PM_{10} mass: yearly average contributions were 9–22 %, 17–22 %, 29–34 % and 21–35 % of PM_{10} levels measured at rural, urban-industrial, urban and traffic sites, respectively. For $PM_{2.5}$, road dust contributions were lower but the same pattern is shown: 7 %, 6–16 %, 11–31 % and 21–31 % respectively. Road dust contributions were generally higher during the dry season. Negative trends across sampling years were found at traffic and urban sites, while positive trends at industrial-harbor area of Algeciras Bay (Gibraltar Strait) probably due to local traffic peculiarities. Relatively to vehicle exhaust, road dust contributions generally decrease with a lower rate and less significantly.

The observed deposited mass loadings of road dust were attributed, in average, to carbonaceous source (i.e. tire wear and bitumen) for 52 %, to a road wear/mineral source for 35 % and to brake wear for 13 % which supports decision making to develop preventive measures to reduce road dust build-up.

Supplementary material related to this article is available online at http://www.atmos-chem-phys-discuss.net/13/31933/2013/acpd-13-31933-2013-supplement.pdf.

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Table 1. Details of PM monitoring sites and sampling periods.

Monitoring site	Province	Type of station	Latitude	Longitude	Altitude (m a.s.l.)	Period	Number of filters
Campillo	Málaga	Rural	37°2′43.27″ N	4°51′41.23″ W		PM_{10} (2010) $PM_{2.5}$(2009)	95
Poblado	Córdoba	Rural	38°6′36″ N	4°55′36″ W		PM_{10} (2010)	21
Lepanto	Córdoba	Urban	37°53′39,1″ N	4°46′5,5″ W	123	PM_{10} (2007–2010) $PM_{2.5}$ (2009–2010)	225
Alcalá de Guadaira	Seville	Urban	37°20′31,4″ N	5°49′59,7″ W	60	PM_{10} (2007–2010) $PM_{2.5}$ (2009)	196
Granada Norte	Granada	Traffic	37°11′50,8′ N	3°36′28′ W	689	PM_{10} (2007–2010) $PM_{2.5}$ (2009–2010)	217
Carranque	Málaga	Traffic	36°43′12,8″ N	4°25′45,8″ W	36	PM_{10} (2007–2010) $PM_{2.5}$ (2009–2010)	222
Príncipes	Seville	Traffic	37°22′35,6″ N	6°0′15,4″ W	8	PM_{10} (2007–2010) $PM_{2.5}$ (2009)	214
Algeciras	Cádiz	Urban Industrial	36°07′50″ N	5°26′51″ W	20	PM_{10} (08/2003–07/2004) $PM_{2.5}$ (08/2003–07/2004)	161
La Línea	Cádiz	Urban Industrial	36°9′34.1"N	5°20′54.39" W	4	PM_{10} (03/2003–2010) $PM_{2.5}$ (03/2003–2010)	632
Los Barrios	Cádiz	Urban Industrial	36°10′31.6" N	5°28′51.2" W	45	PM_{10} (02/2003–2010) $PM_{2.5}$ (02/2005–2010)	455
Puente Mayorga	Cádiz	Urban Industrial	36°10′59.7"N	5°23′11.8" W	8	PM_{10} (09/2004–2010)	258

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Table 2. Road dust composition averaged per road category.

%	Urban roads	Industrial roads	Nearby works	Poor state of pavement
OC	19.2 ± 3.4	15.7 ± 0.2	17.2 ± 3.0	12.7
EC	5.3 ± 2.1	3.2 ± 2.3	4.4 ± 1.6	2.1
Al	2.1 ± 0.6	2.1 ± 0.4	3.2 ± 0.9	4.0
Ca	11.2 ± 3.1	19.7 ± 1.6	12.7 ± 1.6	9.9
Fe	2.5 ± 0.5	2.0 ± 0.6	3.1 ± 1.4	3.1
K	0.7 ± 0.2	0.5 ± 0.1	1.1 ± 0.3	1.5
Mg	1.9 ± 1.3	1.3 ± 0.3	2.6 ± 1.5	0.9
Na	0.3 ± 0.1	0.2 ± 0.1	0.3 ± 0.1	0.3
P	0.1+0.0	0.1 ± 0.0	0.1 ± 0.0	0.1
S	0.4 ± 0.1	0.3 ± 0.2	0.4 ± 0.1	0.2
Cl^-	0.9 ± 1.5	0.1 + 0.0	0.2 ± 0.3	0.0
NO_3^-	0.2 ± 0.1	< LD	0.2 ± 0.1	<LD
SO_4^{2-}	1.0 ± 0.4	< LD	0.9 ± 0.3	0.3
NH_4^+	< LD	< LD	< LD	0.0
$\mu g\,g^{-1}$				
Hg	0.46 ± 0.51	0.42 ± 0.14	0.53 ± 0.28	0.30
Li	17 ± 4	16 ± 4	23 ± 7	26
Sc	4 ± 2	5 ± 0	6 ± 2	7
Ti	1589 ± 355	1638 ± 478	2064 ± 640	2439
V	57 ± 10	74 ± 1	65 ± 6	55
Cr	145 ± 61	682 ± 782	131 ± 40	117
Mn	371 ± 93	626 ± 279	485 ± 261	567
Co	12 ± 3	45 ± 27	12 ± 2	10
Ni	209 ± 135	324 ± 200	164 ± 64	54
Cu	770 ± 266	301 ± 4	702 ± 272	401
Zn	1313 ± 326	1292 ± 545	1141 ± 299	654
Ga	6 ± 2	6 ± 1	8 ± 2	10
Ge	0.40 ± 0.61	<LD	0.68 ± 0.67	0.52
As	19 ± 9	13 ± 5	15 ± 3	18
Se	3 ± 2	6 ± 0	4 ± 0	3
Rb	30 ± 10	28 ± 2	47 ± 12	61
Sr	178 ± 76	203 ± 19	220 ± 83	178
Y	10 ± 4	11 ± 2	11 ± 5	16
Zr	152 ± 57	88 ± 8	167 ± 23	160
Nb	12 ± 3	30 ± 24	17 ± 5	23
Mo	46 ± 75	56 ± 8	62 ± 5	28
Cd	1.0 ± 0.4	6.8 ± 3.5	0.7 ± 0.1	0.5
Sn	107 ± 28	25 ± 15	115 ± 58	81
Sb	54 ± 18	15 ± 7	59 ± 32	40
Cs	1 ± 1	2 ± 0	4 ± 1	5
Ba	755 ± 276	387 ± 194	931 ± 279	533
La	11 ± 4	15 ± 2	15 ± 6	25
Ce	23 ± 9	22 ± 3	32 ± 14	53
Hf	5 ± 1	2 ± 0	5 ± 1	4
W	9 ± 19	5 ± 7	3 ± 3	6
Tl	0.00 ± 0.00	0.31 ± 0.44	0.00 ± 0.00	0.00
Pb	117 ± 17	194 ± 4	149 ± 34	126
Bi	4 ± 2	2 ± 0	6 ± 4	3
Th	5 ± 2	4 ± 0	7 ± 3	10
U	4 ± 2	2 ± 1	4 ± 0	3

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Table 3. Average absolute ($\mu g\,m^{-3}$) and relative (%) source contributions to PM_{10} and $PM_{2.5}$ levels.

PM size	Province	Monitoring site	Type of site	PM ($\mu g\,m^{-3}$)	Sources Mineral	Secondary Nitrate	Heavy oil	Vehicle exhaust	Metallurgy	Secondary sulphate	Sea salt	Road dust	Tyre wear	Traffic/ Secondary
PM_{10}	Seville	Príncipes	Traffic	41	6.7 (16 %)	2.0 (5 %)		8.1 (20 %)		4.9 (12 %)	3.6 (9 %)	14.4 (35 %)		
PM_{10}	Granada	Granada Norte	Traffic	44	4.9 (11 %)			8.8 (20 %)		3.9 (9 %)		10.4 (24 %)	3.4 (8 %)	10.9 (25 %)
PM_{10}	Málaga	Carranque	Traffic	43	10.6 (25 %)		3.0 (7 %)	8.0 (19 %)		4.1 (10 %)	6.9 (16 %)	9.0 (21 %)		
PM_{10}	Seville	Alcalá	Urban	41	6.1 (15 %)	2.5 (6 %)		4.7 (12 %)		5.0 (12 %)	4.1 (10 %)	13.9 (34 %)		
PM_{10}	Córdoba	Lepanto	Urban	36	6.0 (17 %)			5.6 (16 %)	3.9 (11 %)	3.9 (11 %)	4.6 (13 %)	10.4 (29 %)		
PM_{10}	Cádiz	Algeciras	Urban industrial	38	5.2 (14 %)	3.1 (8 %)	2.1 (5 %)	6.2 (16 %)	0.2 (< 1 %)	7.5 (20 %)	4.6 (12 %)	6.6 (17 %)		
PM_{10}	Cádiz	Los Barrios	Urban industrial	30	6.6 (22 %)	4.7 (16 %)	1.5 (5 %)	2.9 (10 %)	0.3 (1 %)	4.5 (15 %)	3.3 (11 %)	5.5 (18 %)		
PM_{10}	Cádiz	La Línea	Urban industrial	37	6.8 (19 %)	4.7 (13 %)	2.2 (6 %)	4.9 (13 %)	0.6 (2 %)	5.8 (16 %)	6.2 (17 %)	6.3 (17 %)		
PM_{10}	Cádiz	Puente Mayorga	Urban industrial	39	8.7 (22 %)	4.9 (13 %)	2.4 (6 %)	3.7 (10 %)	0.5 (1 %)	4.7 (12 %)	4.5 (12 %)	8.4 (22 %)		
PM_{10}	Málaga	Campillo	Rural	15	2.6 (17 %)		2.9 (19 %)	3.5 (23 %)		2.3 (15 %)	1.3 (8 %)	1.3 (9 %)		
PM_{10}	Córdoba	Poblado	Rural	18	6.6 (37 %)			0.7 (4 %)	0.5 (3 %)	1.0 (6 %)	0.7 (4 %)	3.9 (22 %)		
$PM_{2.5}$	Seville	Príncipes	Traffic	31	3.7 (12 %)	1.1 (4 %)		5.8 (19 %)		4.8 (16 %)	1.5 (5 %)	9.5 (31 %)		
$PM_{2.5}$	Granada	Granada Norte	Traffic	37	2.3 (6 %)			6.5 (18 %)		1.3 (4 %)		8.2 (22 %)	6.6 (18 %)	9.9 (27 %)
$PM_{2.5}$	Málaga	Carranque	Traffic	23	2.8 (12 %)		2.7 (12 %)	2.8 (12 %)		4.1 (17 %)	2.4 (10 %)	4.8 (21 %)		
$PM_{2.5}$	Seville	Alcalá	Urban	31	2.9 (9 %)	0.9 (3 %)		3.2 (10 %)		5.4 (18 %)	1.5 (5 %)	9.5 (31 %)		
$PM_{2.5}$	Córdoba	Lepanto	Urban	18	1.5 (8 %)			3.8 (22 %)	3.4 (19 %)	3.5 (20 %)	2.1 (12 %)	1.9 (11 %)		
$PM_{2.5}$	Cádiz	Algeciras	Urban industrial	25	0.9 (4 %)	2.9 (12 %)	2.6 (11 %)	5.0 (20 %)	< 0.1 (< 1 %)	6.2 (25 %)	1.5 (6 %)	1.5 (6 %)		
$PM_{2.5}$	Cádiz	Los Barrios	Urban industrial	22	1.6 (7 %)	2.4 (11 %)	2.0 (9 %)	2.3 (10 %)	0.1 (< 1 %)	5.6 (25 %)	2.6 (12 %)	3.6 (16 %)		
$PM_{2.5}$	Cádiz	La Línea	Urban industrial	20	1.1 (5 %)	2.3 (11 %)	2.7 (13 %)	3.5 (17 %)	0.1 (1 %)	6.9 (34 %)	1.9 (9 %)	1.7 (8 %)		
$PM_{2.5}$	Málaga	Campillo	Rural	19	2.4 (12 %)		3.9 (20 %)	3.9 (20 %)		2.7 (14 %)	4.7 (24 %)	1.4 (7 %)		

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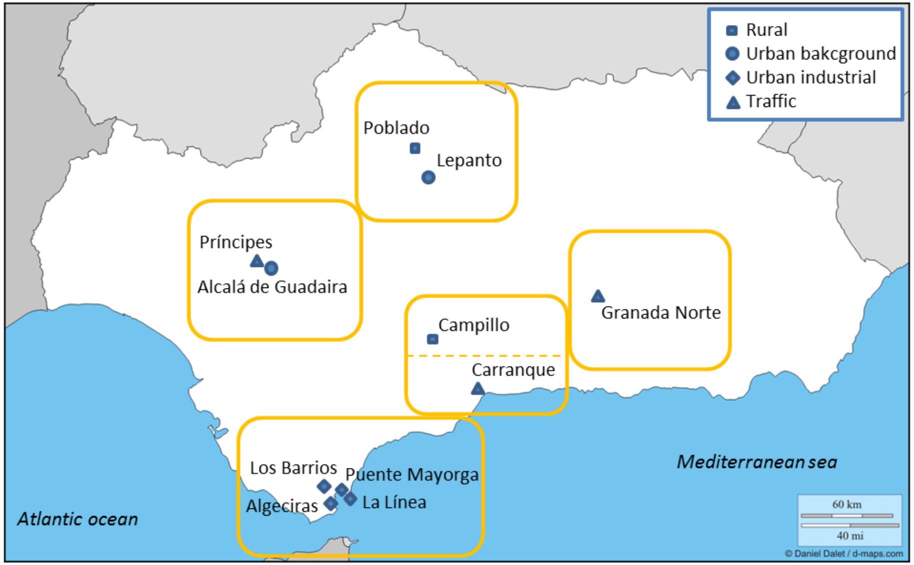

Fig. 1. Map of the 11 monitoring sites (divided in six zones) used for the PM source apportionment study.

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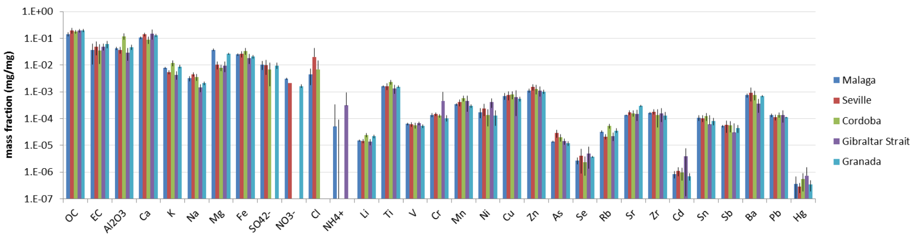

Fig. 2. City-averaged road dust composition at the five cities under study. Error bars indicate standard deviation between four roads.

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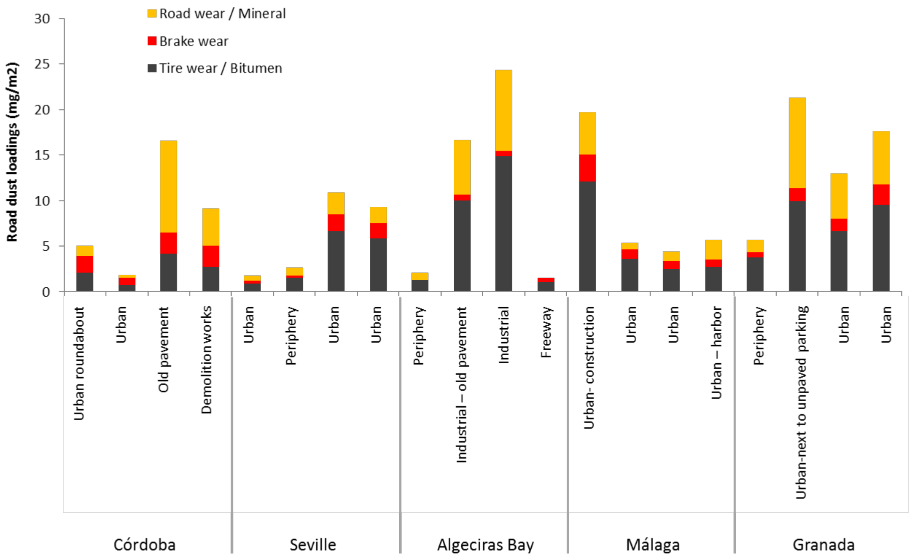

Fig. 3. Absolute contribution (mg m^{-2}) of road dust sources to road dust mass loadings.

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Trends of road dust emissions contributions on ambient PM levels

F. Amato et al.

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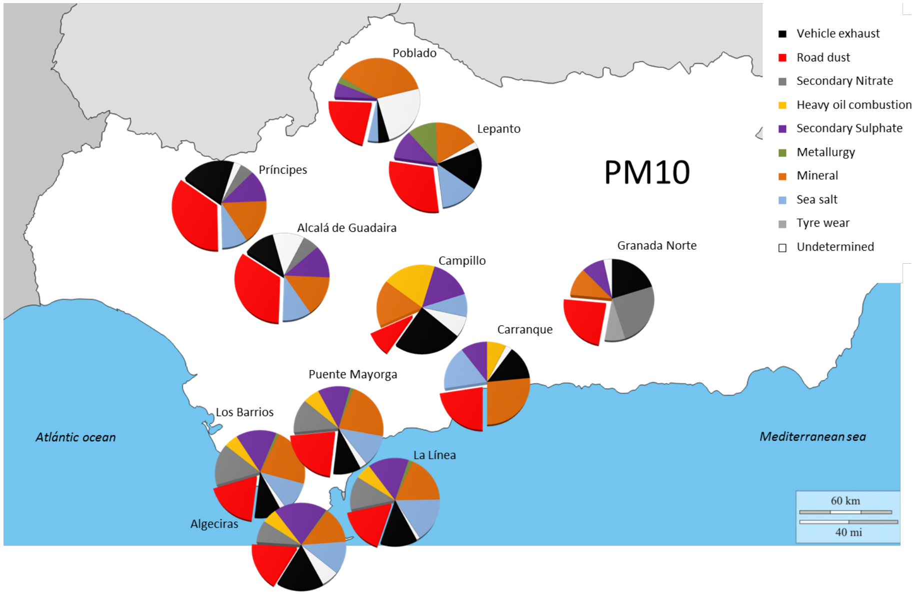

Fig. 4. Pie chart of average source contributions to PM_{10} levels observed at the 11 stations in Andalucia.

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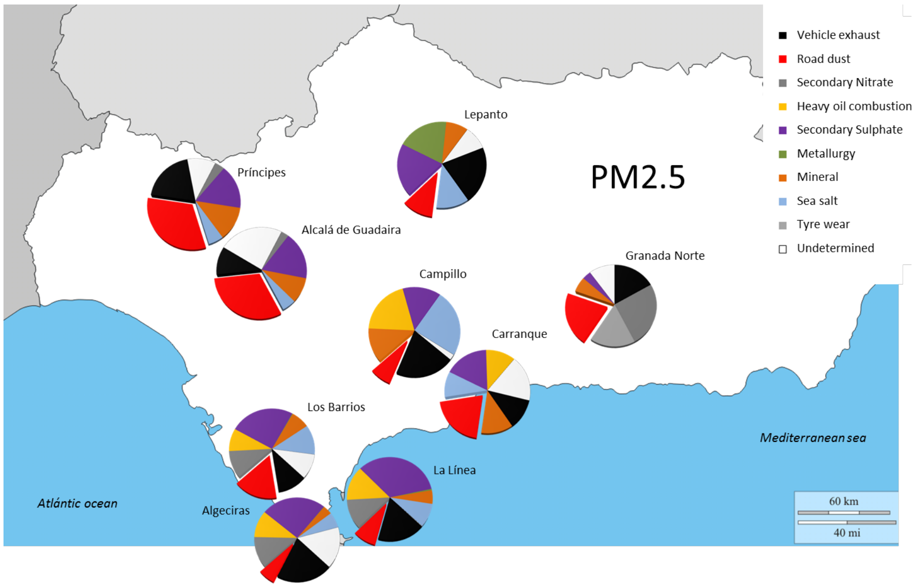

Fig. 5. Pie chart of average source contributions to $PM_{2.5}$ levels observed at the 9 stations in Andalucia.

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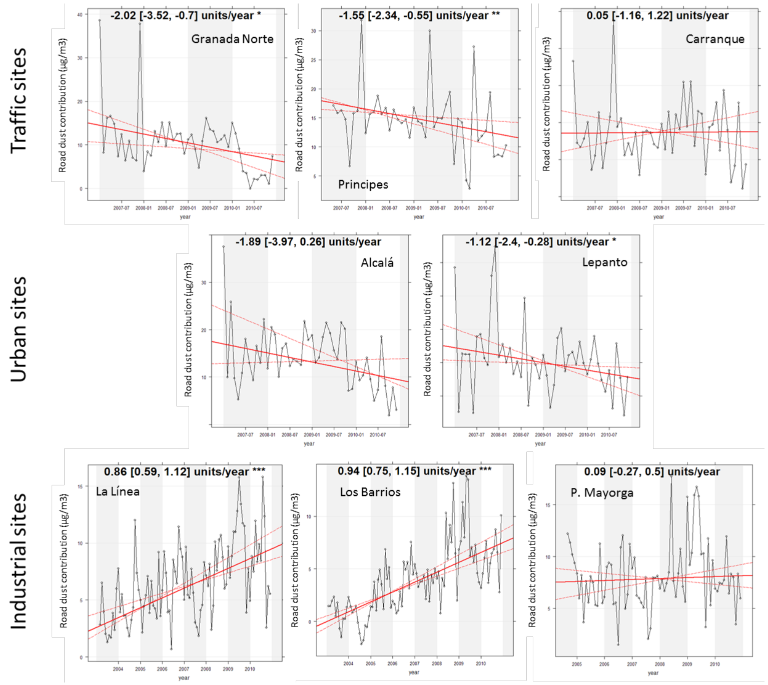

Fig. 6. Temporal trends for road dust contribution in PM_{10} at traffic (Granada, Seville and Malaga), urban background (Seville and Cordoba) and industrial sites (Algeciras Bay). The plots show the deseasonalised monthly mean concentrations. The solid red line shows the trend estimate and the dashed red lines show the 95 % confidence intervals for the trend. The overall trend and the 95 % confidence intervals in the slope (between brackets) are shown at the top as units (µgm-3) per year. The *, ** and *** show that the trend are significant to the 0.05, 0.01 and 0.001 levels, respectively.

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