REVIEW ARTICLE

Atmospheric particulate matter and air quality in the Mediterranean: a review

Sergio Rodríguez · Xavier Querol · Andrés Alastuey · Jesús de la Rosa

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Abstract This article synthesises the results obtained in several projects on atmospheric aerosol (particulate matter – PM) pollution developed during the last years in the Western Mediterranean. It focuses on particulate matter sources and the strategies for suitable monitoring in ambient air. The article has been structured in several sections that give response to the main questions that prompted these studies. After analysing the main PM features in the Mediterranean, a brief review of the main factors differentiating the particulate matter composition with respect to Central Europe is presented. Finally, the suitability of the different metrics or parameters for monitoring ambient air PM levels in different types of environments is discussed.

Keywords Atmospheric aerosol \cdot PM10 \cdot PM2.5 \cdot Saharan dust \cdot Mediterranean \cdot Air quality

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S. Rodríguez (⊠) · X. Querol · A. Alastuey Institute of Earth Sciences "Jaume Almera"-CSIC, Lluis Sole i Sabarís S/N, 08028 Barcelona, Spain e-mail: srodriguez@ija.csic.es

S. Rodríguez · J. de la Rosa Department of Geology, University of Huelva, Campus del Carmen, 21071 Huelva, Spain

S. Rodríguez Izaña Atmospheric Observatory, INM-CSIC, La Marina 20, planta 6, 38071, Santa Cruz de Tenerife, Canary Islands, Spain

Introduction

Atmospheric particulate matter (PM), or simply atmospheric aerosol is a complex mixture of solid and/or liquid substances suspended in an atmospheric gas. Aerosols enter into the atmosphere by several mechanisms including both anthropogenic and natural sources. Organic matter, sulphate, nitrate, ammonium, elemental carbon, mineral dust and sea salt are the main PM contributors ($\sim \mu g/m^3$), whereas some other elements such as As, Cd, Cr, Cu, Ni, P, Pb, V or Zn (among others) are typically found as trace contributors (few or fractions of ng/m^3). Aerosol's diameter varies between a few nanometers (nm) and tens of microns (μ m), depending on the sources, formation mechanisms and 'age' of the particles. Human activities alter the natural PM composition and increase its levels in ambient air several times more than the natural background concentrations. Aerosols influence some of the processes related to climate, e.g. precipitation patters, radiation transfer, among others and cause harmful effects on human health. Owing to these adverse effects linked to aerosol pollution, standards for ambient air PM10 (PM<10 μ m) levels exist within the EU territory (EU Directive 1999/30/EC). Because of the uncertainties about the levels, sources, composition and spatial and temporal distribution of PM across Europe when the Directive 1999/30/EC was prepared, the suitability of these limit values has been recently subject to review (II Position Paper on PM, 2004; a draft of a new PM air quality directed was presented in September 2005). This has resulted in a modification of the PM10 limit values and also a proposal to establish limit values for PM2.5 (PM<2.5 μ m).

This article synthesises the results obtained in several research projects developed since the end of the 1990s, with the aim of contributing to reduce the above-cited uncertainties around PM. Most of the data discussed here were obtained

in Spain and focused on the PM source contribution (e.g. natural vs. anthropogenic) and in the strategies for a suitable PM monitoring in ambient air. Among other reasons, these studies on the strategies for a proper PM monitoring are necessary in the Mediterranean area as the region is frequently affected by natural mineral dust transport from North Africa. These studies have mostly been multidisciplinary, including in-situ chemical and physical characterisation of PM, meteorological analysis and interpretation of satellite remote sensing aerosols observations. Since the first results, it was observed that the processes affecting the levels and composition of PM in Spain were significantly different to those described for other regions of Central and Northern Europe and the factors prompting these differences were specific of the Mediterranean basin. In this paper, we present a synthesis of the most important results on (1) the main features of PM in the Western Mediterranean, (2) the differences with respect to PM features in Central and Northern Europe and (3) the implications for a suitable air quality monitoring in order to avoid natural interferences and trace the PM sources. Although not all Mediterranean regions are properly covered, these results allow obtaining a comprehensive picture of the Mediterranean aerosol behaviour in the western regions. This is a summary of 'results, conclusions and implications', for details on 'methods and specificities,' references to previous publications will be provided in the manuscript.

Experimental

The studies on PM10 and PM2.5 source contribution were performed in different types of environments and climatic regions of Spain. Each sampling campaign lasted at least 1 year, and the sampling was simultaneously performed at least at two–five sites (trying to cover rural, urban and industrial sites). Following this method, a total of about 20 sampling sites were covered across Spain since 1999. These sites were distributed across the different regions of the mainland Spain and the Canary and Balearic Islands. In addition to PM sampling in fixed stations, several short campaigns (days-to-weeks) were also performed around specific PM sources.

The sampling of PM10 and PM2.5 was performed on quartz filters by using high-volume samplers. In each sample,

PM concentration was determined by gravimetric standard techniques and the chemical composition was determined by several techniques, including the ion chromatography or capillary electrophoresis (for sulphate, nitrate and chloride), specific electrode (for ammonium), thermal analysis (for carbon) and ICP-AES and ICP-MS (for a total of 32 elements). This method allows to quantify the content of secondary inorganic compounds-SCI (non-sea salt-SO₄²⁻, NO₃⁻, NH₄⁺), organic matter and elemental carbon (OM + EC), mineral dust $(Al_2O_3 + SiO_2 + CO_3^{2-} + Ti + Fe + Mg + Sr + Mn)$, sea salt (sea salt- SO_4^{2-} + Na + Cl) and trace elements (Cr, Cu, Pb, V, Zn, Co, As and Cd, among others) in the PM10 and PM2.5 samples. Details on the indirect determinations (for SiO_2 and CO_3^{2-}) and laboratory procedures for the sampling conditioning, extraction in ultra pure water and acid digestion are provided by Querol et al. (2001a, b).

This database on PM composition was complemented with in-situ continuous measurements of gaseous pollutants, PM concentrations and meteorology, and interpreted using several tools and methods, such as receptor modelling (for the source contribution identification and contribution), meteorological charts, air back trajectories, satellite observations (TOMS) and modelling forecast for dust production and transport (mostly SKIRON and more recently DREAM as well). For the sake of brevity, only the data from some sites and some examples will be shown here.

Results and discussion

The manuscript is structured in several sections which gives response to the main questions that allows to know the main features of PM in the Western Mediterranean.

The Mediterranean context

The objective of this first section is to describe the 'context' in which the PM anthropogenic emissions take place and the 'key points' that prompt some of the PM features that will be described in the following sections.

The factors affecting the dispersion, transport, levels and composition of PM experience significant variation across Europe (Fig. 1). The areas at the north of the Alps and





μ g/m ³		PM	Mineral dust	OM + EC	SIC	nss-SO4 ²⁻	NO ₃ -	$\mathrm{NH_4^+}$
Rural	PM10	15-20	2–5	2–5	5-8	3–4	1–3	1
	PM2.5	8-12	1-2	2-5	4–5	3–4	≤ 1	≤ 1
Urban background	PM10	30-45	8-12	5-10	6-11	3–5	1.5-4	1-2
	PM2.5	20-30	3	5-10	5–9	3–5	1-2	1-2
compounds $(NO_3^- + Hot spot)$ nss-SO ₄ ²⁻ + NH ₄ ⁺). Hot spots (urban and industrial)	PM10	45-50	13-15	10-18	8-15	3–6	4-6	2-3
	PM2.5	30–35	4–5	10-18	8-13	3–6	2–4	2–3
	μg/m ³ Rural Urban background Hot spot	μg/m³RuralPM10 PM2.5Urban backgroundPM10 PM2.5Hot spotPM10 PM2.5	$\begin{array}{c c} \mu g/m^3 & PM \\ \hline Rural & PM10 & 15-20 \\ PM2.5 & 8-12 \\ Urban background & PM10 & 30-45 \\ PM2.5 & 20-30 \\ Hot spot & PM10 & 45-50 \\ PM2.5 & 30-35 \end{array}$	$\begin{array}{c cccc} \mu g/m^3 & PM & Mineral dust \\ \hline Rural & PM10 & 15-20 & 2-5 \\ PM2.5 & 8-12 & 1-2 \\ Urban background & PM10 & 30-45 & 8-12 \\ PM2.5 & 20-30 & 3 \\ Hot spot & PM10 & 45-50 & 13-15 \\ PM2.5 & 30-35 & 4-5 \\ \end{array}$	$\begin{array}{c ccccc} \mu g/m^3 & PM & Mineral dust & OM + EC \\ \hline Rural & PM10 & 15-20 & 2-5 & 2-5 \\ PM2.5 & 8-12 & 1-2 & 2-5 \\ Urban background & PM10 & 30-45 & 8-12 & 5-10 \\ PM2.5 & 20-30 & 3 & 5-10 \\ Hot spot & PM10 & 45-50 & 13-15 & 10-18 \\ PM2.5 & 30-35 & 4-5 & 10-18 \\ \end{array}$	$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	$\begin{array}{c ccccccccccccccccccccccccccccccccccc$

Pyrenees (e.g. UK, north and western regions of France and Germany) are characterised by a relatively flat terrain, mean westerly winds and frequent passages of cold fronts and depressions resulting in rain. These conditions favour the frequent renovation of air masses. In contrast, the areas surrounding the Mediterranean basin are characterised by an abrupt topography, when coupled with the characteristic synoptic scale patterns, results in low mean wind speeds (i.e. 'synoptic component' of wind) that hinder the air mass renovations and favour the accumulation of PM in the surrounds of emission regions (Fig. 1a and b). In Eastern Spain and Northern Italy, the concatenation of 'low wind speed events and abrupt entries of air masses' prompts the concatenation of 'pollution episodes and clean air events', respectively. Moreover, Eastern Spain is the European region with the lowest precipitation rates (Fig. 1c). Because rain is the main mechanism for atmospheric aerosol removal, this has an important consequence: 'lower precipitation rates imply higher residence time and consequently higher background PM levels'. These low rainfall rates coupled with the 'weak advections of air masses and the frequent air masses recirculation episodes' result in frequent regional pollution events that increases the aerosol's residence time in the region and gives rise to the high PM background levels recorded in summer in Eastern Spain. This specific setting of the Western Mediterranean (including the high radiation intensity) enhances the formation of secondary aerosols and ozone.

In addition to the above features affecting pollutants dispersion and transport, there are some other features favouring the occurrence of higher ambient air concentrations of mineral dust in the Mediterranean basin than in Central and Northern Europe. First, the Mediterranean is frequently affected by African dust outbreaks. Second, because of the low precipitation rates in the Western Mediterranean (1) soils exhibit a poor vegetal coverage in such a way that natural resuspension processes contribute to increase the mineral dust contributions to ambient PM levels, (2) mineral dust accumulation on roads is favoured, and consequently road traffic resuspension is enhanced with respect to cities located in more rainy regions, where washout of road dust reduced resuspension. Examples and details are provided below. See prior publications for the details on meteorology (Millán et al. 1997, 2002) and its influence on PM levels (Rodríguez et al. 2001, 2003).

PM levels, sources and chemical profiles

Annual mean PM levels across the country increase from 15 to 50 μ g/m³ for PM10 and from 8 to 35 μ g/m³ for PM2.5 from the rural to the urban-industrial hot spots (Table 1). OM + EC and SIC exhibits similar concentrations in PM10 and PM2.5, whereas mineral dust concentrations in PM10 are two-four times higher than those in PM2.5 (Table 1). Because of this coarse nature of mineral dust, its contribution to PM2.5 (15–20%) is much lower than that to PM10 (mostly 25–30%) levels. Later, it will be shown how these loads of mineral dust both in PM10 and PM2.5 are much higher in Spain than in Central Europe. The main contributors to the PM increase from the rural to urban sites are mineral dust and OM + EC: the hot-spots/rural ratios for mineral dust, OM + EC and SIC are within the ranges 3.0-6.5, 3.5-5.0 and 1.5-2.0 for PM10 and 2.5-4.0, 3.5-5.0 and 2.0-2.5 for PM2.5, respectively. This relative low increase in the SIC compounds is mainly due to the relatively high background levels of ammonium sulphate in the regional scale. Sea salt contributions are into the ranges 0.8–3 μ g/m³ in PM10 (except in the Canary Islands, where it is $\sim 10 \ \mu g/m^3$ in PM10) and 0.5–1 μ g/m³ in PM2.5.

The application of receptor modelling techniques (e.g. principal components analysis (PCA) for the source identification) to the PM chemistry data base has allowed identifying the chemical profile of the main PM sources across the country (see examples for specific sites in Querol et al. 2001a b; Rodríguez et al. 2004). The most frequently observed as 'first factor' (accounting for the highest variance in the PCA) represents the mineral dust blowing in ambient air (chemical profile containing Al, Ca, Ti, Sr, Fe, K, Mn and Mg). This mineral dust is a mixture of African dust, natural-resuspension and anthropogenic dust (including road, construction and demolition dust) contributions. As second factor it is usually found a chemical profile representative of fuel combustion mostly for power generation, containing nss-SO₄²⁻, V, Ni and at some sites NO_3^- as well (depending on the partitioning of nitrate between the particulate and gas phases, which is controlled by temperature). This chemical profile also contains NH_4^+ because sulphate and a fraction of nitrate occurs as ammonium sulphate and ammonium nitrate, respectively. The following frequently observed chemical profile contains NO₃⁻, OM, EC, Zn and Fig. 2 a-d Monthly values of PM10, organic matter + elemental carbon, nitrate and sulphate in (◆) urban BARCELONA-IJA (NE), (□) urban HUELVA (SW), (▲) urban LLODIO (North), (○) rural MONTSENY (NE) urban sites, e-h Daily mineral concentrations of mineral dust in PM10 and PM2.5 in urban sites during 2001



Cu associated with vehicle exhaust, and ammonium owing to the occurrence of ammonium nitrate. Vehicles exhaust (last chemical profile) and road dust resuspension (a fraction of the first chemical profile) account for the previously discussed strong increase in OM + EC and mineral dust from the rural to the urban sites (Table 1). Finally, sea salt (Na, Cl) contributions are also identified. At some specific sites, other sources such as biomass burning, crude oil refinery or ceramic manufacture were also identified but are not discussed here because these have not a countrywide influence (e.g. Rodríguez et al., 2004; Querol et al., 2002).

PM events and seasonal evolution

Time series analysis of PM levels on the base of the meteorology, allowed identifying and classifying the PM episodes in Eastern Spain as follows: local urban PM pollution events (mostly in the cold season), regional PM pollution episodes (warm season), Atlantic/Northern clean air advections events and African dust outbreaks (Rodríguez et al. 2003, 2005). Clean air events are mostly associated with abrupt entries of Atlantic or Northern air masses into the Mediterranean basin, whereas pollution episodes are prompted by the 'calm air' conditions dominating between these Atlantic air masses entries and the reduced dispersion conditions linked to the topography and regional meteorology. The chemical composition and the source contribution during these events changes significantly as a consequence of the marked seasonal evolution of several PM components.

PM constituents exhibit different seasonal evolutions depending on their formation mechanism and thermodynamics properties (Fig. 2). Both in urban and rural areas nitrate exhibits a seasonal evolution with a maximum in winter owing to the enhanced stability of ammonium nitrate at low temperatures (Fig. 2a; Adams et al. 1999). In urban areas, OM + ECexhibit a maximum in winter (Fig. 2b; owing to the accumulation of PM in the reduced urban boundary layer and the enhanced condensation of semi-volatile organic compounds at low temperatures), whereas in rural areas OM + EC does not show a well-defined pattern across the country. Both in urban and rural areas, sulphate concentrations maximise in summer due to the enhanced SO₂ to sulphate conversion rates because of the involvement of photochemistry (Fig. 2c). Owing to these opposite seasonal trends of the main PM components, bulk PM10 and PM2.5 levels in urban areas do not show a well-defined seasonal evolution (as that of its components; Fig. 2d). By contrast, 'rural PM10' concentrations ('regional background') and the difference between 'urban and rural PM10' levels (equivalent to the 'local urban contribution') tend to exhibit well-defined seasonal trends (Fig. 3). 'Rural PM10' concentrations exhibit a maximum in summer, mainly because the enhanced contribution of ammonium sulphate and mineral dust (natural resuspension on arid soils and African dust events; Fig. 3a). In contrast, the 'local urban contribution' (difference between 'urban and



Fig. 3 Monthly mean concentrations of 'rural regional' and 'urban local' (urban–rural) contributions to PM10 and its most important contributors in Eastern Spain (deduced from two rural sites, MON-AGREGA and MONTSENY, and two urban sites L'HOSPITALET and BARCELONA-IJA)

rural PM10' levels) maximises in winter owing to the strong contribution of ammonium nitrate and OM, and secondly by road mineral dust (Fig. 3b). Thus, ammonium sulphate is the most important PM contributor during summer 'regional PM episodes', whereas local urban ammonium nitrate, OM and road mineral dust are the most important contributors to PM during 'local urban pollution episodes' characteristics of the cold season.

African dust events

Desert dust outbreaks over the Mediterranean occur in a synoptic scale, affecting large areas of Southern Europe under well characterised meteorological scenarios (see studies on meteorology and modelling in Rodríguez et al. 2001 and on satellite observations in Rodríguez et al., 2003). Daily mean concentrations of mineral dust during these events are typically within the ranges 10–15 μ g/m³ in PM2.5 and 15– 60 μ g/m³ in PM10 in mainland Spain and 10–40 μ g/m³ in PM2.5 and 30–100 μ g/m³ in PM10 in the Canary Islands (e.g. Fig. 2e–h). The magnitude of these mineral dust concentrations in Spain agrees with other observations (in TSP) in Sardinia (Molinaroli et al. 1993) and Turkey (Kubilay et al. 2000). During extreme events mineral dust concentrations are as high as 100 μ g/m³ in PM10 in mainland Spain, up to 600 μ g/m³ in PM10 and 65 μ g/m³ in PM2.5 in the Canary Islands (Viana et al. 2002), 135 μ g/m³ in PM10 and 65 μ g/m³ in PM2.5 in Portugal (Almeida et al. 2005) and 190 μ g/m³ in PM10 and 90 μ g/m³ in PM2.5 in Crete (EMEP, 2004), have been recorded. Owing to this African dust transport results in PM10 episodes, this is an agent interfering with the monitoring of PM and air quality. Around 10 African dust events per year (2–4 days duration each) occur in many parts of the Mediterranean, contributing to the increase in the number of exceedances of the EU daily limit value for PM10.

European PM composition: Mediterranean versus Northern regions

This database on PM composition in Spain has contributed to several compilations of PM composition across Europe (e.g. Van Dingenen et al. 2004; Putaud et al. 2004; Querol et al. 2004a,b). The results support our first observations, pointing to the high load of mineral dust as the main feature of PM in the Western Mediterranean. Querol et al. (2004a) obtained that the annual mean mineral dust concentrations in PM10 across Spain are several times higher than those typically recorded in Central Europe (Austria, Germany, Switzerland, The Netherlands and the UK). The ratios of the mineral dust contents in PM Spain with respect to Central Europe at rural background (RB), urban background (UB) and kerbsides (KS) sites are 4.0 at RB and 2.5 at UB and KS in PM10, and 1.5-2.0 at RB, 2.5-5.0 at UB and 3.0-4.0 at KS in PM2.5. It is important to highlight how the most important differences in terms of PM10 are recorded at the rural sites (ratio = 4), whereas in terms of PM2.5 are recorded at KS (ratio = 3-4). As previously stated, this high load of mineral dust in the Western Mediterranean is the result of the frequent African dust events and the strong mineral dust resuspension, owing to natural causes at rural sites and by traffic forced resuspension of road, construction and demolition dust at urban sites. This traffic forced resuspension is strongly enhanced by the dust accumulation in streets and roads because of the low precipitation rates in the Western Mediterranean (scarce self cleaning; Fig. 1c). These observations are also supported by another study on PM2.5 across Europe (Götschi et al. 2005). These data compilations also support other peculiarities of the Western Mediterranean area: the $nss-SO_4^{2-}$ and rural PM concentrations seasonal evolution with a maximum in summer.

PM monitoring parameters

As previously stated, PM2.5 is mostly constituted by combustion-derived compounds (except ammonium). The contributions of mineral dust and sea salt are much higher in PM10 than in PM2.5 because the coarse nature (>2.5 μ m) of these two components and the <2.5 μ m size of the combustion-derived compounds (Table 1). Given this





segregation in the particle size according to the composition, the selection of the most suitable PM parameter varies as a function of the PM source to be monitored. Thus, in areas affected by industrial activities releasing precursors of secondary aerosols (both organic and inorganic), PM1 and PM2.5 are suitable parameters but not PM10 if there are significant levels of mineral dust as in the Western Mediterranean. See in Fig. 4a1 the very good correlation between PM1 and SO₂ recorded during fumigations of a power-plant's plume in Eastern Spain. In this region, PM10 and PM 1-10 do not 'detect' the fumigations because of the important contributions of mineral dust in the 1–10 μ m fraction due to natural wind resuspension on the arid soils (Fig. 4a2). In contrast, PM10 is a suitable parameter for monitoring PM pollution around industrial activities involving the manipulation of mineral dust (e.g. ceramic, cement or brick production, mostly linked to fugitive emissions) because the mass of dust is mainly in the >2.5 μ m fraction. Urban PM emissions require the monitoring of both PM10 and PM2.5, owing to PM2.5 accounts for the exhaust emissions and PM10 includes the road dust contributions as well.

Because of this link between PM sources and particle size, the PM2.5/PM10 ratio experiences significant variations (Table 2), showing values (1) as high as 0.7–0.8 during fumigations of secondary aerosol precursors plumes, (2) as low as 0.1–0.4 during resuspension of mineral dust and fugitives emission in industrial activities involving the manipulation of mineral dust as raw matter (e.g. mining activities and cement, brick or ceramic production), and (3) 0.4–0.7 in urban areas. The mean PM2.5/PM10 ratios in ambient air of urban areas experiences a northward gradient across the country with values 0.4–0.5 in the Canary Islands, 0.5–0.6 in the Southern Iberian regions and 0.6–0.7 in the northern regions. In very polluted urban and industrial areas, this ratio is reaches 0.7 across the country. Querol et al. (2001) and Alastuey et al. (2004) provide more examples.

The current PM monitoring metrics are all based on the mass-concentrations (mass/volume) of PM below certain cut

 Table 2
 Mean ratios if PM in several size fractions around PM sources and in ambient air in Eastern Spain

	PM2.5/PM10	PM1/PM10
Fumigations of plumes:		
Power plants $(SO_2)^a$	0.8	0.5
Petrochemistry ^b	0.7	0.3
Biomass burning ^b	0.5	0.2
Fugitive emissions in ^b :		
Mining activities	0.1	< 0.1
Raw matter transport	0.3	< 0.1
Atomisers ceramic industry	0.3	0.1
Resuspension forced by ^c		
Wind on arid soil	0.4	0.2
Crop cultivation	0.2	0.1
Ambient air in:		
Barcelona kerbside		
5Pth	0.3	0.1
25Pth	0.4	0.2
50Pth	0.5	0.3
75Pth	0.6	0.4
98Pth	0.7	0.6
Rural top of mountain ^c		
5Pth	0.6	0.4
25Pth	0.7	0.6
50Pth	0.8	0.7
75Pth	0.9	0.8
Rural arid-ebro basin ^d	0.5	0.2

Pth: percentile

^aTeruel power plant; ^bIndustrial areas of Castelló; ^cMontseny; ^dEbro basin

size (e.g. PM10 and PM2.5). However, because of the use of more advanced technologies during the last decade, the 'aerosol mass' emissions have been significantly reduced, whereas significant emissions in terms of 'aerosol number' are taking place nowadays, mainly from road traffic exhaust (e.g. Longley et al. 2005). Because the main contributors to the aerosol number concentrations (ultrafine PM, i.e. <0.1 μ m) are not the most important contributors to aerosol mass, the number of fine particles *N* and its associated PM2.5 mass concentrations are 'not equivalent' (non-univocally related) parameters. See in Fig. 4b1 how PM2.5 correlates very well with the number of particles coarser than 0.4 μ m $(N>0.4 \ \mu m)$ and do not show a strong relationship with the ultrafine particles number concentration $N < 0.1 \ \mu$ m. Given that $N < 0.1 \,\mu$ m is being emitted in significant amounts nowadays, that this ultrafine aerosol has adverse effects on human heatlh (Wichmann et al. 2000) and that they are not properly accounted for in PM2.5, the monitoring of the ultrafine number concentration should also be carried out in air quality networks, at least in urban areas ,because road traffic is the most important ultrafine PM source. However, more research is needed in order to have a comprehensive picture of the relationship between the number and mass concentrations. A recent study has shown that this relationship may experience significant variations depending on the proximity to the sources, 'age' of the particles and dominant microphysical processes (Rodríguez et al., 2005).

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