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Submicron particle concentration and particle size distribution at urban and rural areas in the surroundings of building materials industries in central Spain

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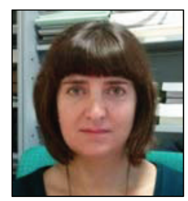
ABSTRACT

Particle number concentrations and size distribution of atmospheric aerosol particles were measured in the range 6–560 nm by a fast mobility particle sizer in Toledo, Spain, and in the outskirts of nearby small villages within the area of influence of a cement factory, quarries and ceramic industries. Several measurement campaigns have been carried out in different days in Toledo (April, May and July) and one campaign in "Comarca de la Sagra" in July. In each campaign, measurement of 10–15 min has been achieved in several different points. In addition, a campaign of continuous measurements has been carried out in Toledo. The average number concentration in Toledo are consistent with other urban measurements, with lower values during the night–time ranging from $3x10^3$ to $1x10^4$ particles/cm^3 and higher average levels during daytime ranging from $5x10^3$ to $4x10^4$ particles/cm^3 depending on the measurement site and date. The measurement sites show similar background profiles of the particle size distributions with two clear particle modes centered in the sizes ranges 10–15 and 40–50 nm. Road traffic is postulated as the main source of submicron particles, giving lower level of particles during the summer time. Results from continuous measurements have been analyzed. On the other hand, the low total concentrations obtained in the "Comarca de la Sagra" campaign, ranging from $1.5x10^3$ to $3.1x10^3$ particles/cm^3 with an average value of $2.3x10^3$ particles/cm^3, show a negligible influence of the different building materials industries on the total submicron particle concentration in the surrounding area and in Toledo.

Keywords: Aerosol, pollution, size distribution, submicron particles, urban air



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1. Introduction

Suspended particulate matter is an important pollutant for which regulation in most countries includes only PM_{10} particles. The more recent ambient air quality legislations tend also to include fine particles ($PM_{2.5}$) (U.S. EPA, 2012). In contrast, no regulatory action has been adopted yet concerning the potentially more harmful particles, submicron and ultrafine particles (UFPs, with diameters below 100 nm). UFPs account for 80–90% of the total particle number concentration in urban air (Mejia et al., 2008), and exposure to them have very important health influence on blood and different organs (e.g. Delfino et al., 2005; Araujo and Nel, 2009; Weichenthal, 2012; Patterson et al., 2014) needing more epidemiological and toxicological evidences (Kumar et al., 2010).

The main natural sources of nanoparticles include forests, oceans and atmospheric formation (Seinfeld and Pandis, 2006), in urban areas vehicle exhausts, especially form diesel engines, and new particle formation in ambient air (Harris and Maricq, 2001; Keogh et al., 2009; Pey et al., 2009). In both natural and urban air, the reactions of OH and ozone are involved in the formation of secondary organic aerosols. In this sense, the reported levels of surface ozone in different areas (Notario et al., 2012; Notario et al., 2013a) may correlate with the number concentration of ultrafine particles. Other anthropogenic source of UFPs is manufacture of new nanomaterials with local effects in the areas of production or use, cement factories, etc. (Andujar et al., 2009; Kumar et al., 2010). Thus for example, emissions of fugitive dust from cement where the fraction of particles with diameter sizes <1 µm may amount up to ~10% in weight (Baroutian et al., 2006), or areas with a large number of new buildings in construction, may contribute to the sub–micrometric fraction of suspended matter. Concerning the surrounding cities, these particles are much more reactive than those from natural mineral dust (Kunal et al., 2012), being necessary additional studies to assess the impact on air quality (Abdul–Wahab, 2006).

In Spain, a large portion of population lives in small cities: 47% in villages of <50 000 inhabitants and 24% in small cities from 50 000 to 200 000 inhabitants. Only 29% of population lives in great cities (>200 000 inhabitants) (INE, 2013). In the USA for example, the population living in towns with 50 000–200 000 inhabitants (approximately 54 million people) over–exceeds the population in cities over 500 000 inhabitants (Approximately 40 million people) (Census, 2010). The atmospheric plume in big cities may be quite different from that of smaller towns. For large cities, the contribution from the outside may be very small due to the huge urban emissions over a large territory what may induce the general behavior. In contrast, small cities may be more sensitive to the effect of given emissions in the surrounding areas. Thus, the need to characterize aerosol particle size distribution at different locations, especially in small cities, is clear. Several recent works conducted in Spain includes rural sites (Notario et al., 2013b), industrial and urban environments (Fernandez–Camacho et al., 2012), urban sites, or great cities outdoor and indoor environments (Reche et al., 2014). In the present work, we report the first field study concerning the levels of UFPs particles in central Spain. The campaign has been conducted within the city of Toledo to evaluate the emissions from vehicles and in the region "Comarca de la Sagra" in the north–east of Toledo, a rural zone with relatively low population but with potential emissions from a cement factory and several ceramic industries and quarries.

One of our targets is to contribute to the knowledge of air pollution due to sub–micrometric particles in small cities and to the evaluation of the effects of the surrounding area. Also, back trajectories were calculated in order to determine the origins and pathways of the air masses affecting the studied region. The results may be very useful for environmental authorities to identify the areas potentially affected by this important and harmful pollutant and to enable clinical studies.

2. Observation Sites, Equipment and Campaigns

2.1. Observation sites

The Historic City of Toledo, declared World Heritage in 1986, is one of the most important touristic cities in Spain. It is placed in the Tagus valley in the centre of the Iberian Peninsula in the region of Castilla–La Mancha (39°52'N, 4°1'W) at 529 m above sea level, approximately 70 km south from Madrid (Figure 1a). The city has some 82 000 inhabitants. The old part of the city is located on the right bank of the Tagus River, on a hill a hundred feet above it, which encloses its base, forming a sharp bend. Since the mid–nineteenth century new buildings are growing outside the walls, and now Toledo has a relatively dispersed population, with open spaces from the core neighborhoods. The residential district "Santa Maria Benquerencia" is situated at east on the right bank of the Tagus River, separated from the city centre about 6 km (Figure 1a). Toledo is surrounded by a ring road that allows road–traffic to connect the city with motorways.

At northeast from the city of Toledo we find the "Comarca de la Sagra", a region of ~700 km^2 within this province which extends from Toledo to the western edge of the province of Madrid (Figure 1b). The basement of docile clay has favored the ceramic industry engaged in building materials and the nature of the soil has allowed the exploitation of quarries and cement industries. All these industries are potential sources of emissions of several pollutants including particles of different sizes. According to the annual emission inventory of 2010 performed by the Spanish Government, for Toledo the following emissions were obtained (in tons per year): NMVOCs 6 291, NO_X 10 388, total particles 950 and SO_X 10 612 (PRTR–Espana, 2010). Meteorologically, this region has very hot and sunny weather during summer, dry and cold winters, with very low wind speed throughout the year in addition to frequent temperature inversions. This city experiences numerous daylight hours (about 3 000) and high solar radiation levels, making this area suitable for surface oxidant formation (including ozone).

2.2. Equipment

Particle number concentration and size distribution were obtained by a fast mobility particle sizer (FMPS model 3091, TSI Inc.) (Notario et al., 2013b). Measurements were made in the range 6–560 nm with 32 size channels (16 channels per decade of size). A high sample flow rate (10 L/min) helps to minimize particle sampling losses due to diffusion, and operation at ambient pressure prevents evaporation of volatile and semi–volatile particles. The technique is based on particle electrical mobility. The FMPS consists of a particle charger, a classification column, and a series of detection electrometers. In this instrument, a unipolar charger produces a defined number of chargers predictable on the particles which enter the instrument through a PM_1 (particulate matter of particles with an aerodynamic diameter dp<1 μm) cut–off. The particles which flow through the region of different electrical fields are repelled by the voltage from the central column. When the particles hit the outer cylinder, consisting of multiple cylindrical rings, the particles create a current measured by electrometers.

The FMPS was situated in the cargo area of a sport utility vehicle and was powered by a high–capacity battery linked to a DC–AC converter. The scan time of one cycle is 1 second and most of measurements were carried out for ~10–15 min with the car stopped. Then, the FMPS was transported to the next measurement point for about the same time, so on to complete the study. Sampling was tested at three different heights; for this purpose, the FMPS was attached to a ½'' inner diameter copper tubing which could be extended to heights of 125 cm, 225 cm and 275 cm above the ground. No differences were found for the 3 heights, what shows good mixing conditions and a negligible effect from the ground. Most of the measurements were carried out at 225 cm.

In order to determine the origins and pathways of the air masses affecting the measurement sites involved in this study, back trajectories were computed using the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model Version 4 developed by the NOAA's Air Resources Laboratory (ARL) (Draxler et al., 2009). The GDAS input meteorological files have a spatial resolution of 1x1 and 24 levels of vertical resolution. The three–dimensional kinematic back trajectories were calculated using the vertical wind component provided by the meteorological model. In order to understand the behavior of the air masses circulating in the planetary boundary layer (PBL), these trajectories were calculated at a 100 m height.

2.3. Campaigns

In Toledo, several campaigns have been carried out. The first campaign was conducted on April, 12th 2011 in different places of the city from 9:00 to 14:30 local times (UTC+2). Figure 1a shows a map of these measurement sites marked from (1) to (9).

Two pedestrian sites, in the outskirts (1) and the center of the city (2), were chosen to enable the comparison with sites with usual traffic. Point (1) is located beside the banks of the Tagus River inside the University Campus in a pedestrian area. Point (2) corresponds to the only measurement carried out within the walls of the old city, where traffic in general is prohibited except to residents, loading and unloading vehicles, taxis and official cars. Thus, in points (1) and (2) there should not be in principle significant direct vehicle emissions but they are surrounded by neighborhoods with road traffic. The rest of sites (3–9) are typical of vehicle exhausts emissions.

Sites (8) and (9) are located in the neighborhood of "Santa Maria de Benquerencia" at 6 km east from the city centre, with an industrial park in (8) where there are many small companies or a sewage treatment, and the residential area (9) where approximately 30% of Toledo's population lives.

A second set of measurements was conducted on 26–May–2011. Since slight differences had been found previously in the different measurement sites, no new measurements were carried