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# Short-term effects of ultrafine particles on daily mortality by primary vehicle exhaust versus secondary origin in three Spanish cities

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## ABSTRACT

*Background:* Evidence on the short-term effects of ultrafine particles (with diameter $<$ 100 nm, UFP) on health is still inconsistent. New particles in ambient urban air are the result of direct emissions and also the formation of secondary UFP from gaseous precursors. We segregated UFP into these two components and investigated their impact on daily mortality in three Spanish cities affected by different sources of air pollution.

*Methods:* We separated the UFP using a method based on the high correlation between black carbon (BC) and particle number concentration (N). The first component accounts for aerosol constituents emitted by vehicle exhaust (N1) and the second for the photochemical new particle formation enhancements (N2). We applied city-specific Poisson regression models, adjusting for long-term trends, temperature and population dynamics.

*Results:* Mean BC levels were higher in Barcelona and Tenerife (1.8 and 1.2 $\mu g \cdot m^{-3}$, respectively) than in Huelva (0.8 $\mu g \cdot m^{-3}$). While mean UFP concentrations were similar in the three cities, from which N1 was 40% in Barcelona, 46% in Santa Cruz de Tenerife, and 27% in Huelva. We observed an association with N1 and daily mortality in Barcelona, by increasing approximately 1.5% between lags 0 and 2, per an interquartile increase (IQR) of 3277 $cm^{-3}$, but not with N2. A similar pattern was found in Santa Cruz de Tenerife, although none of the associations were significant. Conversely, in the industrial city of Huelva mortality was associated with N2 at lag 0, by increasing 3.9% per an IQR of 12,032$\cdot cm^{-3}$.

*Conclusion:* The pattern and origin of UFP determines their short-term effect on human health. BC is possibly the better parameter to evaluate the health effects of particulate vehicle exhaust emissions, although in areas influenced by domestic solid fuel combustion this should also be taken into account.

## 1. Introduction

A large number of epidemiological studies have reported an association between particulate matter with an aerodynamic diameter $< 10\,\mu m$ ($PM_{10}$) or $< 2.5\,\mu m$ ($PM_{2.5}$) and daily mortality (Katsouyanni et al., 2009; Pérez et al., 2009; Samoli et al., 2013). Moreover, toxicological findings suggest stronger effects of ultrafine particles (with diameter $<$ 100 nm, UFP) compared with $PM_{2.5}$ or filtered air, exhibited significantly larger early atherosclerotic lesions in mice (Araujo et al., 2008). Because of the differences in deposition and the potential for translocation as well as their huge active surface, effects of UFP might be at least partly independent from those of larger particles, such as $PM_{2.5}$ and $PM_{10}$ (HEI, 2013). But only a few epidemiological studies investigated the association UFP and health outcomes. Some found an association (Atkinson et al., 2010) but others reported inconclusive results (HEI, 2013). More recently, the UFIREG study conducted in Central Europe (Lanzinger et al., 2016) and the UF&Health in Nordic and Mediterranean cities (Stafoggia et al., 2017),

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found weak evidence of an association between UFP and daily mortality. These inconclusive results might be attributed to the different UFP origins of the cities involved in the studies, or even to the different origin of UFP for different days in a city.

New particles in ambient urban air arise mostly from primary, or direct particulate, vehicle exhaust emissions (Charron and Harrison, 2003) and from in situ, urban or regional nucleation (Brines et al., 2015; Ma and Birmili, 2015; Hofman et al., 2016; Salma et al., 2016; Kontkanen et al., 2017). In addition to these two major sources, other sources such as industrial, airport, shipping and domestic emissions can also contribute to increase levels of both primary and secondary UFP (Keuken et al., 2015; Buonanno and Morawska, 2015; Kecorius et al., 2016). Previous studies have already indicated that vehicle exhaust represents the main primary emission source in urban areas, mainly releasing UFP in terms of number concentration (N, in $cm^{-3}$) (Wehner and Wiedensohler, 2003; Gidhagen et al., 2005; Hofman et al., 2016).

Rodríguez and Cuevas (2007) suggested a method based on the high correlation between black carbon (BC, also emitted mostly from vehicle exhausts in urban environments of Europe (Reche et al., 2011a)) and N, which is typically observed in urban ambient air, to segregate the contributions to ambient air N into two groups ($N = N1 + N2$). The component N1 accounts for those UFPs directly emitted in the particle phase and those components nucleating immediately after emission. The component N2 accounts for mostly nucleation during the dilution and cooling of the vehicle exhaust emissions or by other urban and regional nucleation processes (Brines et al., 2015), but also due to contributions of local or transported low BC-bearing UFPs from different sources (Cheung et al., 2011). The composition and related features of N1 and N2 might thus be very different. The first being mostly made of diesel soot (high hydrocarbon and BC UFPs) and the second of organic and inorganic secondary UFP components. However, the effects of both segregated components, N1 and N2, on human heath have not been studied yet.

The objective of this study is to assess the short-term association between the two segregated components of ambient UFP and daily mortality, from same day of the death up to two days before, in three Spanish cities with urban environments affected by different sources of air pollution.

## 2. Methods

### 2.1. Study areas and monitoring sites

Data were collected from three Spanish cities covering a wide range of climate and emission patterns (Fig. 1).

Barcelona, located in the North-western Mediterranean Basin, has 1.61 million inhabitants (3.2 million if the metropolitan area is considered) and is characterised by a dense road traffic network, also influenced by industrial and shipping emissions. Barcelona is a small city (104 $km^2$) and bears one of the highest population and vehicle densities of Europe, and its fleet is characterised by a large proportion of diesel cars and a large number of motorbikes (Table 1). The harbour of Barcelona has the highest number of cruise ships for tourists in Spain ($4 \times 10^6$ passengers in 2016), which results in a significant focus of emissions of atmospheric pollutants that are transported across the city by the sea breeze (Pérez et al., 2016). The city, and its ambient PM, is also affected by the emissions from industrial estates located in its surroundings (Amato et al., 2010). Measurements took place in an urban background monitoring station at the Institute of Environmental Assessment and Water Research (IDAEA-CSIC), close to one of the largest road traffic arterial of the city (230 m to the North of the Diagonal Avenue, $7 \times 10^4$ vehicle/day).

Huelva is a city in the South-western Spain with a population of around 0.15 million. Besides the typical urban emissions, also with a large proportion of diesel cars (Table 1), Huelva is influenced by the particulate and gaseous emissions from two large industrial estates located close to the harbour at the south of the city. The "Punta del Sebo" industrial state includes the second copper smelter in Europe and relatively high ambient levels of $SO_2$, As, Cd, Cu, Pb, Zn and Bi, among others, have been identified in the area (Fernández-Camacho et al., 2012). Moreover, this park also has phosphoric acid production plants, which contributes with emissions of $NH_3$, phosphate, $PO_4^{-3}$, HF and HCl (Alastuey et al., 2006). The main emissions in the other industrial park, Nuevo Puerto, come from a petroleum refinery: volatile hydrocarbons, $SO_2$, $NO_x$, $NH_3$, Ni and V. The sea-to-land breeze results in these industrial/shipping emissions being transported to the city of Huelva. The Campus El Carmen monitoring station is part of the air quality network of the Autonomous Government of Andalusia, operated in collaboration with the University of Huelva. The closest roads lie about 500 and 1000 m apart.

Santa Cruz de Tenerife has 0.20 million inhabitants. It is located in the Canary Islands, at the bottom of the southern slope of the Anaga ridge and the eastern slope of the North-East to South-West ridge crossing the island. Its topographic characteristics protects the city form the trade winds (North-East) that blow over the ocean (Guerra et al., 2004). The city is mainly affected by road traffic emissions from a fleet with low proportion of diesel cars (Table 1), emissions from the ships and cargo operations taking place in the harbour, and from the oil refinery located in the southern side of the city (Rodríguez et al., 2008; González et al., 2011; González and Rodríguez, 2013). The measurements were carried out at the urban background station of the Santa Cruz Observatory from the Meteorological State Agency of Spain (AEMET). It is located on a large avenue that runs parallel to the coast and carries intense road traffic load. The location of the station and the prevalent winds might favour an impact of this industrial plant in the monitored air quality (González and Rodríguez, 2013).

Additional details on ambient measurements are reported in the Table S1 in the Supplementary material.

### 2.2. Data collection

For each city, data were collected on daily counts of all-cause mortality, excluding deaths from external causes (International Classification of Diseases, 9th and 10th Revisions: ICD9 codes 001–799 and ICD10 codes A00–R99) for all ages.

Hourly data on UFP, measured as N, as well as BC mass concentrations were available from long monitoring campaigns conducted in each city, within the frame of the EPAU project (Rodríguez et al., 2008), at least covering three consecutive years; Barcelona from 2009 to 2014, Huelva from 2008 to 2010 and Santa Cruz de Tenerife from 2008 to 2012. Although as stated above UFP are those with a size < 100 nm, when measuring N it is well accepted that around 80% of the N from 1 to 1000 nm, falls in the UFP size range. That is why we use indistinctively N and UFP. Additional details on instrumentation used for N and BC measurements are reported in Table 1. We have to notice that the lower cut size diameter is lower in the CPC models used in Huelva and Santa Cruz de Tenerife (2.5 nm) than in Barcelona (5 nm). Consequently, absolute concentrations are not directly comparable and relatively lower counts might be expected for Barcelona. At the three sites, BC was measured in $PM_{10}$. This is not expected to cause major differences since most BC is expected to fall in the $PM_1$ fraction.

Finally, time series data on daily mean temperature (°C) were used to control for the potential confounding effects of weather.

### 2.3. The segregation method

Depending on their source, UFP can be classified into (i) primary, which include directly-emitted particles, and (ii) secondary, when newly formed into the atmosphere from gas-phase precursors. As previously stated in Rodríguez and Cuevas (2007) developed a methodology that allows the quantification of the primary (N1, mainly from road traffic) and secondary (N2) contributions to N. Since this method