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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·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 μm (PM_{10}) or < 2.5 μm ($PM_{2.5}$) 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 (4×10^6 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, 7×10^4 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

Fig. 1. Geographical location of the three study areas (Barcelona, Huelva, and Santa Cruz de Tenerife).

is based on the high correlation between BC and N in urban atmospheres this procedure requires collocated BC and N measurements in urban air quality monitoring sites. We assume that N = N1 + N2, where N1 is estimated by applying a semi-empirical scaling factor to BC concentrations, as N1 = S1 × BC (Rodríguez and Cuevas, 2007). The S1 slope corresponds to the 1st percentile of the N/BC ratio recorded during the morning rush hour (07:00–09:00 LT). Specifically, it corresponds to the minimum (N) # cm^{-3} directly emitted into the atmosphere per each ng m^{-3} BC from vehicle exhaust (Rodríguez and Cuevas, 2007). Hourly averages of N and BC were used for the determination of S1 and N1, and subsequently of N2.

For each city, a different S1 was determined per each year, due to different fleet composition and meteorological conditions during traffic rush hours, and to changes in the instrumentations. The city-specific S1 slopes used for the determination of the contribution of N1 and N2 are reported in Table S2 in the Supplementary material.

According to this methodology, N1 corresponds to the minimum primary emission of vehicle exhaust, while N2 accounts for (i) newly formed secondary particles originated by nucleation from gaseous precursors, from road traffic or other urban sources, (ii) low BC-bearing primary particles from a source other than road traffic such as biomass burning, residential and biogenic emissions, and (iii) particles transported with the air mass (Reche et al., 2011b; Cheung et al., 2011). This methodology was also successfully applied in European cities (Reche et al., 2011a; Hama et al., 2017), and in an Asian megacity and boreal forest (Kulmala et al., 2016).

2.4. Statistical analysis

The association between N, N1 and N2 and daily mortality were investigated using Poisson regression models allowing for over-dispersion. We used natural cubic splines as a smoothing function for time, with six degrees of freedom per year to control for seasonality. To control for weather, we included two smooth terms of temperature in order to better account for both high and low temperatures at different reference lags. For high temperatures, we calculated the average temperature on the current and previous day (lag 0–1) and fit a natural spline with three degrees of freedom on the lagged variable only for days on which the lag 0–1 temperature was higher than the median annual temperature for the city as a whole. Similarly, we adjusted for low temperatures by fitting a natural spline with two degrees of freedom for the average temperature on previous 6 days (lag 1–6) only for days on which the lag 1–6 temperature was below the median annual value for the city (Samoli et al., 2013; Stafoggia et al., 2017). We also fitted indicator variables for the day of the week, public holidays and the population decrease during the summer vacation period (Samoli et al., 2013; Stafoggia et al., 2017).

We firstly investigated short-term effects of N using single day lags

Table 1
Description of the city characteristics, vehicle fleet (in brackets % of diesel), site location and instrumentation used for N and BC measurements.

	Barcelona	Huelva	Tenerife
City characteristics			
Population* (hab.)	1,608,746	145,648	203,585
Surface (km^2)	102.1	151.3	150.5
Mean temperature (sd)	16.7 °C (6.2)	20.2 °C (5.9)	22.1 °C (3.8)
Vehicle fleet			
Motorbike	284,407 (0.2%)	12,993 (0.6%)	19,726 (0.5%)
Passenger and LDV	617,175 (42.7%)	73,070 (55.5%)	122,137 (23.6%)
Bus	2523 (89.3%)	138 (99.3%)	1259 (99.2%)
Heavy duty vehicles	43,114 (93.6%)	5625 (96.8%)	16,787 (87.2%)
Other	6953 (60.7%)	796 (62.1%)	1282 (77.8%)
Monitoring sites			
Location	41° 23′ 05″N, 02° 07′ 09″E; 68 m a.s.l.	37° 15′ 0″N, 6° 57′ 0″W; 54 m a.s.l.	28° 29′ 20″N, 16° 18′ 33″W; 52 m a.s.l.
Type	Urban background	Urban industrial	Urban background
Instrumentation			
N	WCPC 3785 (TSI Inc.) $N_{5–1000}$	CPC 3776 (TSI Inc.) $N_{2.5–1000}$	CPC 3776 (TSI Inc.) $N_{2.5–1000}$
BC	MAAP (Thermo ESM Andersen) with PM_{10} inlet	MAAP (Thermo ESM Andersen) with PM_{10} inlet	MAAP (Thermo ESM Andersen) with PM_1 inlet

LVD: Light Duty Vehicles.
sd: standard deviation.
WCPC: water condensation particle counter, CPC: condensation particle counter, MAAP: multi-angle absorption photometer.

models, from the same day to the death (lag 0) up to two days before (lag 2). Next, we estimated the effects of N1 and N2 fitting both components simultaneously for the same lags. We did not include distributed lag structures in our models as the missing pattern of the data prohibits the interpretation of combined lags.

We also performed sensitivity analysis on the time-trend adjustments to check the robustness of the main results by replacing the spline of time trend with a three-way interaction between year, month, and day of the week, an approach previously shown to be equivalent to the time-stratified case-crossover design (Levy et al., 2001). However, because missing data might alter the matching between case days and control days, we also applied a simplified version using a two-way interaction between year and month instead, jointly with indicator variables for day of the week. Since we acknowledge that different patterns and distributions in the missing data across cities might introduce some bias to the effect estimates, these sensitivity analyses should allow detection and quantification of such bias, if it exists (Stafoggia et al., 2017).

All analyses were done using Stata statistical software, release 14 (StataCorp, college Station, TX, 2015). All results are reported as percentage increase in mortality risk (%IR) and corresponding 95% confidence interval (95% CI) relative to an increase equal to city-specific interquartile range in particulate number concentration.

3. Results

Particle number concentration (N) was moderately correlated with BC in Barcelona and Santa Cruz de Tenerife, but poorly in Huelva (Supplemental material, Table S2). Descriptive data on daily levels of N and BC are reported in Table 2. In spite of the differences in lower size detection limits (> 5 nm for Barcelona and > 2.5 nm for the other sites), the three cities reported similar mean levels of N while the highest BC was recorded in Barcelona. The proportion of days with missing values for both, daily N and BC data, were highly variable across cities (28% for Barcelona, 55% for Huelva and 47% for Santa Cruz de Tenerife). The N2 contribution to N prevails over N1 in all three-cities. However, the component N1 reached a larger contribution to N in Barcelona and Santa Cruz de Tenerife (40% and 46%, respectively) than in Huelva (27%), while the component N2 was much larger in Huelva (73%). This is in agreement with the road traffic/industrial UFP contributions documented at the three sites according to their respective emission patterns. Fig. 2 shows how the hourly distribution of component N1 coincides with the maximum traffic rush hours, in the mornings and afternoons, while the N2 component has its maximum value at noon, when photochemical nucleation is expected to be at its maximum. The daily mean number of deaths was 42.4 in Barcelona, ranging from 17 to 78, and similar in Huelva and Santa Cruz de Tenerife, 6.3 (0 to 15) and 7.2 (0 to 20), respectively. Temperature displayed a north-to-south increasing gradient.

Table 3 reports the results of the short-term association between daily mortality and an increase in interquartile range of city-specific particle number concentration, at different lags. In none of the three cities was the daily mortality associated with N for any of the lags evaluated. In contrast, daily mortality was associated with an IQR increase of the component N1 (3277 cm^{-3}) in Barcelona at same day of death, one and two days later, increasing the risk by 1.65% (95% CI = [0.74, 2.52]), 1.52% (0.61, 2.44) and 1.39% (0.47, 2.33), respectively. The component N2 was not associated with daily mortality in Barcelona. A similar pattern was found in Santa Cruz de Tenerife, showing larger effects of N1 than for N2 at lags 0 and 1, although none of the associations was statistically significant. Conversely, in Huelva mortality was associated with an IQR increase of 12,032 cm^{-3} of N2 at same day of death, increasing the risk by 3.95% (0.10, 7.95), whereas the component N1 was not related with.

We also explore longer lagged effects, up to one week before the death, but they were not statistically significant for any component. The results of the sensitivity analyses on time trend adjustment via time-stratified case-crossover design were almost identical to those obtained in the main approach, supporting the long-term and seasonal time trend adjustment in our main models (see Tables S3 and S4 in the Supplementary material).

4. Discussion

Using data from three Spanish cities, with urban environments affected by different sources of air pollution, we reported an association between short-term exposure to segregated components of ambient UFP and daily mortality. We found immediate effects of the minimum primary emission of vehicle exhaust (N1) in Barcelona and also of secondary UFP and low BC-primary UFP from a source other than traffic in Huelva (N2), while no statistically significant effects were found for Santa Cruz de Tenerife for any of the components.

Both BC and N concentration ranges obtained in this study cover most of the urban background concentrations in EU cities. Thus, 1.26 10^4, 1.68 10^4 and 1.42 10^4 cm^{-3} and 1.8, 0.8 and 1.2 μg BC m^{-3} were obtained as average concentrations for Barcelona, Huelva and Santa Cruz de Tenerife, whereas Samoli et al. (2016) reported median N (> 3 nm) of 1.21 10^4 cm^{-3} for North Kensington urban background site in London; Reche et al. (2011a) measured 1.21–1.71 10^4 cm^{-3} and 0.8–1.8 μg·m^{-3} BC for a selection of urban background European sites; and Stafoggia et al. (2017) reported 0.7–1.9 10^4 cm^{-3} for 7 out of 8 European cities studied (range of N: 4–10 nm lower limit; 100–3000 nm upper one).

The lower N concentrations reported for Barcelona might be the result of the coarser lower cut size diameter of the CPC model used compared with the other two cities (> 5 nm vs > 2.5 nm). However, the high influence of large industrial SO_2 emission sources in Huelva may account for the higher UFP concentrations, since the oxidation of SO_2 into H_2SO_4 is driving nucleation of UFP (Kulmala et al., 2004). This is further supported by the higher prevalence of N2 (secondary; 73%) over N1 (primary-traffic) in Huelva. On the other hand, the higher BC levels measured at Barcelona are consistent with its much larger vehicle

Table 2
Descriptive of daily particulate number concentration (N and N1/N2 in cm^{-3}), black carbon (BC in $\mu g \cdot m^{-3}$) in the three study areas.

City	Mean	(sd)	Percentile distribution				
			P5	P25	P50	P75	P95
Barcelona							
N	12,607.7	(5088.5)	5587.7	8799.3	11,859.5	15,669.8	22,124.4
N1	4974.9	(2848.3)	1607.9	2948.0	4332.7	6225.3	10,672.1
N2	7656.0	(4262.3)	1164.6	4617.6	7145.3	10,475.3	15,608.7
BC	1.8	(1.1)	0.6	1.1	1.6	2.4	4.0
Huelva							
N	16,751.8	(12,787.0)	2867.3	8105.6	13,162.4	22,142.3	42,872.2
N1	4514.6	(2522.1)	1717.5	2736.7	4020.3	5496.2	9578.3
N2	12,288.9	(11,729.0)	0.0	4263.4	8776.7	16,296.3	36,786.9
BC	0.8	(0.5)	0.3	0.5	0.7	1.0	1.7
Tenerife							
N	14,150.5	(9130.8)	3072.4	7005.3	12,361.8	19,338.2	30,867.2
N1	6593.1	(4212.8)	1559.4	3499.4	5626.7	8841.0	14,880.1
N2	7598.2	(6387.6)	730.5	2979.2	5973.4	10,563.4	20,353.9
BC	1.2	(0.8)	0.3	0.7	1.1	1.7	2.9

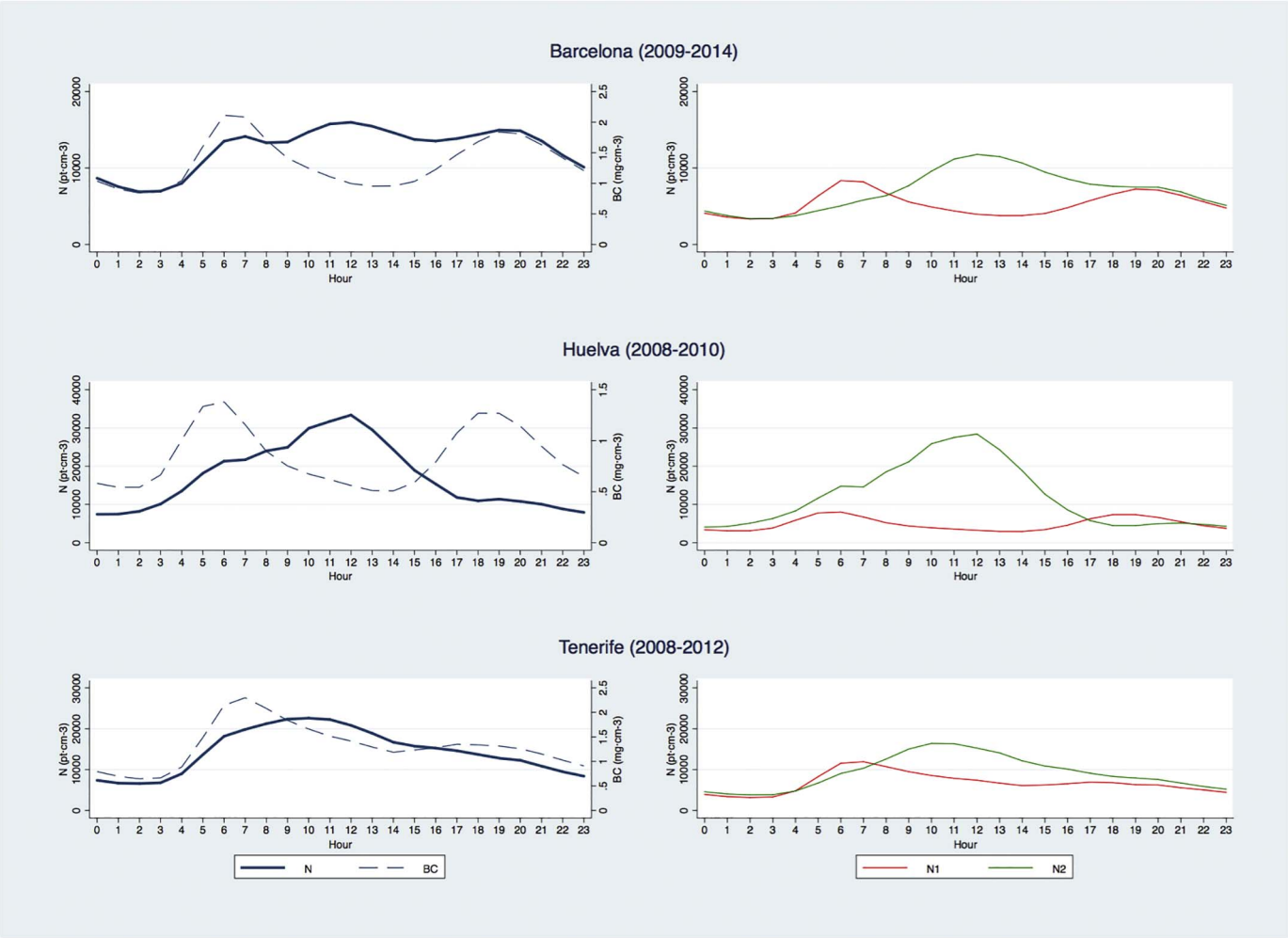

Fig. 2. Average hourly distribution of N and N1/N2 components (in cm^{-3}) and black carbon (BC in $\mu g \cdot m^{-3}$) in the three study areas.

density.

According to the process yielding to increased concentrations of N1 and N2, their composition will markedly differ. Thus, in the case of N1, particles will be made mostly of diesel BC-rich soot particles (with high levels of hydrocarbons) with a mode of 30–40 nm according to cluster analysis performed for UFP in Barcelona (Dall'osto et al., 2013; Brines et al., 2015). However, N2 is expected to be mostly made up of nucleation UFP, with a smaller proportion of low BC primary and secondary UFP from shipping at Barcelona and Santa Cruz de Tenerife with two large harbours and sea breeze prevalence at noon. Thus, N2 is expected to be made mostly by < 20 nm particles with a proportion of highly soluble species generated initially by nucleation of 1–3 nm particles, and followed by growth up to the above size, or even coarser ones. According to Kulmala et al. (2004) it is the system SO_2-NH_3-H_2O

Table 3
Association between daily mortality and N and N1/N2, from single (N) and two-pollutant (N1 and N2) models, in the three study areas: percentage increases of mortality risk (%IR), and 95% CIs, per interquartile range (IQR) in cm^{-3} in particulate number concentration at lags of 0, 1 and 2 days.

City	Lag	N			N1			N2		
		IQR	%IR	(95% CI)	IQR	%IR	(95% CI)	IQR	%IR	(95% CI)
Barcelona	0	6870.5	0.30	(−0.87, 1.49)	3277.4	1.63	(0.74, 2.52)	5857.8	−0.67	(−1.86, 0.54)
	1		0.97	(−0.23, 2.18)		1.52	(0.61, 2.44)		−0.43	(−1.68, 0.84)
	2		0.78	(−0.45, 2.02)		1.39	(0.47, 2.33)		−0.60	(−1.88, 0.69)
Huelva	0	14,036.7	3.14	(−0.92, 7.36)	2759.5	−1.66	(−5.38, 2.22)	12,032.9	3.95	(0.10, 7.95)
	1		0.78	(−3.30, 5.03)		−1.10	(−4.81, 2.75)		1.09	(−2.79, 5.13)
	2		−0.98	(−5.10, 3.33)		1.31	(−2.43, 5.18)		−1.94	(−5.77, 2.06)
Tenerife	0	12,332.9	2.04	(−1.27, 5.46)	5341.6	1.73	(−1.56, 5.13)	7584.2	0.63	(−2.42, 3.77)
	1		0.86	(−2.47, 4.30)		2.07	(−1.59, 5.86)		−0.14	(−3.27, 3.10)
	2		−0.93	(−4.24, 2.51)		−1.05	(−4.78, 2.84)		0.32	(−3.04, 3.79)

that creates the first ion clusters giving rise to nucleation of H_2SO_4, $(NH_4)_2SO_4$, and NH_4HSO_4, although the presence of VOCs favours the generation of the clusters. The subsequent growth is also favoured by condensation of the above species, as well as by condensation of VOC-derived PM. The main SO_2 source in Barcelona is shipping. As previously stated, the N2 composition in the case of Huelva, would also be influenced by industrial emissions and hence have a differing chemical composition from the other two cities. Several studies pointed to a high toxicity of the oxidised organic substances, which are major constituents of secondary organic aerosols (SOA) (Mauderly and Chow, 2008; Biswas et al., 2009; Saffari et al., 2015). The sources of SOA and their composition might importantly determine their toxicity (Lund et al., 2013). The literature on the toxicology and health effects of the organic aerosols is mostly focused on the traffic-sourced ones (Saffari et al., 2015; Win-Shwe et al., 2015). For instance, SOA from traffic emissions increase their oxidative potential as they age (higher photo-oxidation (Saffari et al., 2015)). To the knowledge of the authors, no studies have focused on the health effects of the exposure to secondary aerosols derived from industrial plants, such as the high SO_2 and As emitting smelter or the petrochemical plant from Huelva. Further studies are required to explore and disentangle the possible health effects of N1 and N2 (Cassee et al., 2013), and particularly to specific sources of N2.

This higher relevance of secondary newly formed UFP (N2) in Huelva might also be a plausible explanation for the health outcomes found for N2; whereas in the other two cities, the health outcomes were related with primary traffic UFP (N1). At Huelva, the coastal industrial emissions, with high metal and SO_2 (Cu-smelter), as well as NO_2-and VOCs (petrochemical plant) loads, are transported inland by the sea breeze and might give rise to high N2 episodes (Fernández-Camacho et al., 2012). At Huelva, high concentrations of N2 are usually associated with high loads of As, Cd, Cu, Pb and Zn linked to industrial emissions, including the second most important copper smelter of Europe, fertilizers plants and an oil refinery (Fernández-Camacho et al., 2012). On the other hand, at the other sites, although shipping emissions might supply SO_2 for N2 episodes, the high road traffic N1 emissions seems to supply a similar proportion of UFP. In Barcelona and Santa Cruz de Tenerife the composition of N2 is probably of a less toxic nature than the secondary UFP arising from Huelva's industry, whereas the higher traffic UFP emissions (N1) might account for the higher mortality found in these cities. At these two last sites N2 has also relevant and peak concentrations, with lower intensity than at Huelva, at midday when sea breeze circulations at Barcelona and Santa Cruz de Tenerife transport SO_2 bearing shipping plumes to the city (González et al., 2011; González and Rodríguez, 2013). This SO_2 supply, at midday with high insolation and relatively low pollution (due to maximal wind speed of this period) probably accounts for the H_2SO_4 driven nucleation episodes (Pérez et al., 2016). However, at the three sites, BC (and consequently N1) peaking is recorded at the traffic rush hours coinciding with breeze changes and calm periods (Reche et al., 2011a).

The estimated proportions of N1 and N2 components that we found with the empirical method are supported by real-time measurements and the collection of filters for posterior chemical characterisation reported by previous studies at the three cities (Rodríguez and Cuevas, 2007, Reche et al., 2011a,b, Fernández-Camacho et al., 2012). Still, we conducted an indirect analysis to further assess for the short-term effects of BC and SO_2, as main tracers of the traffic and industrial components in the three cities (Table S5 in the Supplementary material). As expected, BC was strongly associated with daily mortality in Barcelona and Tenerife, showing close risk estimates than N1, but not in Huelva. Conversely, SO_2 was only associated in Huelva, reporting a similar risk than N2, although it was not statistically significant.

Previous studies conducted in Barcelona already found short-term effects of fine particles (PM_1 and $PM_{2.5}$) on daily mortality (Pérez et al., 2009; Samoli et al., 2013) and results from a multisource model shown that traffic is the most important contributor to the adverse health effects linked to PM (Ostro et al., 2011). BC has also been associated with daily mortality in Barcelona, reporting a 2% increase risk of daily mortality for a rise of 1.4 $\mu g \cdot m^{-3}$ (Ostro et al., 2015), exhibiting much greater toxicity per microgram than generic $PM_{2.5}$. However, in a recent study conducted in eight European cities, UFP was not associated with daily mortality (Stafoggia et al., 2017), even though they reported similar estimates to our study. Within the EMECAM study, NO_2 and SO_2 were significantly associated with daily mortality in Huelva, while PM_{10} was only associated during the cold months of the year (Daponte Codina et al., 1999). But so far, no study has already been done to evaluate the health effects of UFP in Huelva. A study conducted in the Canary Islands did not find relationship between PM and daily mortality in Tenerife (López-Villarubia et al., 2010). However, a cohort study of patients with acute coronary syndromes admitted into a tertiary care hospital in Tenerife, found that exposure to UFP was a precipitating factor for admission for heart failure (Domínguez-Rodríguez et al., 2011) and, more recently, that oxidative stress was associated with BC exposure and predicted major adverse cardiovascular events at 30 days (Dominguez-Rodríguez et al., 2015). Moreover, only few epidemiological studies investigated the association UFP and daily mortality. Atkinson et al. (2010) found an association in London, but more recently multicentre studies in Central Europe (Lanzinger et al., 2016) and Nordic and Mediterranean cities (Stafoggia et al., 2017) did not find consistent effects when different cities were included in these studies. Lastly, Samoli et al. (2016) assessed the UFP effects on daily mortality and hospital admissions in London using the nucleation, traffic, accumulation mode UFP in a region where photochemical nucleation is not having a high influence on urban N annual averages (Reche et al., 2011a). Their findings are broadly consistent for the traffic related source, previously reported in London (Atkinson et al., 2010). Thus, to the knowledge of the authors, our study is the first being conducted to assess in a consistent way the short-term effects on human health of UFP according to the traffic (N1) and photochemical

nucleation (N2) origin, in cities were the later can be an important contributor of UFPs.

Due to their small size, UFP have high deposition efficiency, huge active surface, and the potential for systemic translocation (Kreyling et al., 2006; Kelly and Fussell, 2012). These physical characteristics related to UFP size altogether with its chemical composition imply that the biological pathways and health effects of UFP might be noticeably independent from those of larger particles such as PM_{10} and $PM_{2.5}$ (Brook et al., 2004; Rückerl et al., 2011; HEI, 2013). Multiple biological mechanisms are hypothesized to be behind the adverse health effects on different systems of PM (Brook et al., 2010). Some of these pathways may be especially relevant for UFP, since these particles are not well recognized and cleared by the immune system, have higher biological reactivity and can reach the bloodstream and, by extension, get to different target organs (Oberdörster et al., 2005). Indirectly, UPF may begin a series of responses in lung cells, such as the synthesis of reactive oxygen species, producing tissue oxidative stress in the lungs, blood, remote tissues or at a systemic level (Brook et al., 2010; HEI, 2013) that may lead to inflammatory responses. These inflammatory responses may trigger a change in vascular tone (endothelial dysfunction), detrimental cardiac consequences, and a pro-coagulation state (with thrombus formation, ischemic response, and increase of atherosclerotic lesions (Araujo and Nel, 2009; Chin, 2015). Inflammation also causes changes in the balance of the autonomic nervous system, which is in charge of critical body functions, such originates oxidative DNA damage (Bräuner et al., 2007), which is mutagenic and carcinogenic. Finally, particles may translocate trough the circulatory system to other organs and directly induce effects at vital parts such as the heart, or brain.

It is difficult to translate these N1 and N2 compositional, solubility and size patterns, but it is clear that: i) they might have different health effects; ii) N2 levels, composition and related health effect might vary from site to site depending on the emission and climate patterns; iii) N1 composition in densely traffic urban areas will be probably more similar; iv) the ranges on N1/N2 relation in EU cities will depend mostly on the insolation patterns, given that N2 is driven mostly by photochemistry (Reche et al., 2011a); and v) given the different composition, solubility and size of N1 and N2, evaluating the health effects of N (N1 + N2) in low insolation regions (where N will be mostly governed by N1) might allow evidencing similar effects to that reported for BC, but it will require to perform the analysis independently for N2 and N1 in high insolation regions.

The comparison between cities with different characteristics is one of the major strengths of this study, as these differences in exposure; Barcelona and Santa Cruz de Tenerife being importantly influenced by N1, while in Huelva N2 is the predominant component, which are driven by different sources (traffic, industrial, etc.), allow for the identification of potentially harmful pollutants. Furthermore, our study suffers from the typical limitations of epidemiological time-series studies concerning ambient air exposure accuracy and the small magnitude of associations, although it is relevant in public health from the community general exposition. We also have not addressed for contextual socioeconomic status and other potential modifiers, such as age and sex. However, the study design that would have meant a spatial adjustment had been very different to that conducted in our study (Barceló et al., 2009). Also, from a pathophysiological point of view there it is well known that the elderly group is most vulnerable to the short- term effects of air pollution on health (Aga et al., 2003).

5. Conclusion

Our results show that the measurement of BC is probably a better parameter to evaluate in a homogenous way the health effects of particulate vehicle exhaust emissions, although in areas influenced by domestic solid fuel combustion the contribution of this residential source should also be taken into account. Thus, the patterns and origin of UFP determines their short-term effect what poses a big challenge to epidemiological studies unable to discriminate the UFP by its segregates.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envint.2017.11.015.

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