SUPPLEMENTAL MATERIAL

Exposure to ultrafine particles and respiratory hospitalizations in five European cities.

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DESCRIPTION OF STUDY AREAS

Barcelona

The city of Barcelona, NE Spain, with around 1,600,000 inhabitants in the city centre and over 5,000,000 in the metropolitan area, is located on the Mediterranean coast and geographically constrained by the Collserola mountain range to the west, thus atmospheric dynamics are influenced by the breeze regime. This area is characterized by mild winters, warm summers and prevalent clear sky conditions all year round. Solar radiation is thus intense and precipitations are scarce. In the urban background of Barcelona the main source of atmospheric PM is road traffic, although contributions from industry, regional secondary atmospheric pollutants, construction, and shipping are also relevant. Barcelona has one of the highest car densities in Europe (5800 cars taxed in the city/km^2[1]), with diesel vehicles making up around 47% of the fleet,[2] and one of the most important ports of the Mediterranean. In spite of these adverse facts, PM_{10} and $PM_{2.5}$ mass concentrations have decreased considerably in the last decade. Moreover, different meteorological scenarios can have an impact on the levels of pollutants, such as stagnant anticyclonic conditions, recirculation of air masses, Atlantic air mass advection or African dust outbreaks.

Copenhagen

Copenhagen is a city with about 580,000 inhabitants, an area of 86 km^2 and a population density of 6,700 inhabitants km^{-2} located on Sealand, one of the largest Danish islands. Copenhagen is the capitol of Denmark and about 1,000,000 people live in the greater Copenhagen area. There are only few industrial sites in Copenhagen and thus only little contribution from these sites is expected to affect air pollution in Copenhagen. On the other hand a harbor area is located on the eastern side of Copenhagen directly connected to the city where ships stop but also pass by the city crossing the Øresund Strait. On Amager, an island south of the inner city center the Copenhagen airport is located in a distance of less than 10 km where aircrafts take off and land with high frequency as the airport is distributing passengers to other Scandinavian distributions. The overall terrain is flat and approximately at a few meters above sea level. The climate in Copenhagen can be described as marine or continental depending on the prevailing meteorological situation. In general, westerly winds are dominating the main wind directions.

Helsinki

The city of Helsinki is the capital of Finland, with around 600,000 inhabitants. The Helsinki metropolitan area consists of four cities (Helsinki, Vantaa, Espoo, and Kauniainen) and is by far the biggest and most densely populated area of Finland with about 1 million inhabitants. Population density in certain parts of Helsinki's inner city area is very high, reaching 20,000 inhabitants per km^2, but as a whole Helsinki's population density of 2,741 per km^2 (July 2010) ranks it as quite sparsely populated in comparison to other European capital cities. Helsinki metropolitan area is located on a relatively flat land on the coast of the Gulf of Finland. The area has a humid continental climate. Average temperature varies from -5 °C in February to +18 °C in July. The vast majority of the inhabitants live in the urban areas of the cities, but within the boundaries of these cities there are also suburban and rural areas. The majority of homes are heated with district heat throughout the year, but in some areas residential wood combustion is common. The major sources of PM are traffic, energy production and residential wood combustion.

Rome

Rome is the largest Italian city, with 2.9 million inhabitants in a 1,285 km^2 area. The urban area is divided into five concentric circular zones, corresponding with different levels of urbanization, population density and road traffic. The historical center, which corresponds to the limited traffic zone (LTZ) (55,000 inhabitants over an area of 6 km^2), shows the highest concentration of business activity in Rome. Warm months, lasting in Rome generally from

June to September, are generally characterized by large-scale high-pressure systems. Cold months (November – March) are generally characterized by moderately low temperature, prevailing north wind. Moreover periodically high pressure systems can produce temperature inversions and weak winds, leading to stagnation episodes associated with pollutant accumulation in the lowers layers due to poor dispersion conditions. The main pollutants emission sources are road vehicles exhausts and small-scale combustion units, with thermal capacity < 50 MWth used in the civil sector for heating. Services and commerce are the main working activities, and emission of air pollutants from industries is relatively low compared with those originating from road traffic sources.

Stockholm

The city of Stockholm is the capital of Sweden, with around 900,000 inhabitants on 2014. The metropolitan area of Greater Stockholm has around 2 million inhabitants (2014). Stockholm is situated on the south-central east coast of Sweden, where the freshwater Lake Mälaren flows out into the Baltic Sea. The central parts of the city consist of fourteen islands that are continuous with the Stockholm archipelago. Over 30% of the city area is made up of waterways and another 30% is made up of parks and green spaces. The terrain is flat. Annual precipitation is around 500 – 600 mm and mean temperature around 7 °C. The main local source of PNC and PM_{10} is road traffic. PNC is mainly due to local vehicle exhaust, whereas PM_{10} is mainly from non-exhaust traffic emissions and long-range transport. Road wear is more important for PM_{10} concentrations in Stockholm than in many other countries where the use of studded tires is not so common. High PM_{10} concentrations are found in the inner city in springtime mostly due to particle suspension, as a result of the use of studded tires when roads are free from snow. Road surface wetness is very important for PM_{10} with systematically higher levels during dry conditions as compared to wet, while no significant difference can be observed for PNC.

PARTICLES NUMBER CONCNENTRATION (PNC) MEASUREMENT SITES AND METHODS

An overview about the location of the measurement sites and their characteristics is given in Table S1, the measurement periods and methods are summarized in Table S2.

Barcelona

PNC measurements were carried out in Barcelona at two urban background monitoring sites located in the same area during two periods (July 2005 to January 2009 and from February 2009 to February 2011). The sites have similar characteristics and are named “Institut Jaume Almera” (“IJA”, 41°23'5.01"N, 2° 7'9.00"E) and “Torre Girona” (“TG”, 41°23'22.19"N, 2° 6'59.43"E). Both monitoring sites are similarly influenced by vehicular emissions from one of the main traffic avenues of the city, located at a distance of around 200-400 m from the sites (traffic density of 90000 vehicles/working day[1]). However, the first monitoring site (IJA) seems to be more affected by a nearby parking area. In the urban background of Barcelona the main source of atmospheric PM_x is road traffic, although contributions from industry, regional secondary atmospheric pollutants, construction, and shipping are also relevant. Atmospheric dynamics are driven by the daily evolution of the breeze regime, with a NW wind component during the night and the development of breezes during the day turning progressively from SE to SW direction, with gradually increasing wind speeds reaching maximum levels around noon. PNC measurements in the range 5-1000 nm were carried out by means of a water-based condensation particle counter (TSI WCPC3785).

Copenhagen

The particulate matter measurements were carried out on top of H.C. Ørsteds Institute (12^o33'41" E, 55^o42' 1" N), a building of Copenhagen University located in the centre of

Copenhagen. The sampling height of the inlets were about 22m above ground level. In the direct vicinity of the building is the university campus including a park area and the Copenhagen hospital. In front of the building a major street is located. In general, the area outside the park is characterized by 4^{th} floor buildings and a number of major roads connecting the different districts in Copenhagen that are characterized by residential and shopping areas. The whole area is not subject to industrial emissions but to vehicular emissions from major roads nearby. All in all, the measurement site can be described as urban background. PNC was measured by a custom-built DMPS (Differential Mobility Particle Sizer) using a Vienna type medium DMA and a TSI CPC (Condensation Particle Counter) model 3010. $PM_{2.5}$ and PM_{10} measurements were carried out with a standard TEOM (Tapered Element Oscillating Microbalance) instrument.

Helsinki

In Helsinki, the measurement sites were located on campus area in Kumpula, 5 km northeast of the Helsinki center (Kumpula1). From March 2001 to October 2004 the PNC was measured in the fourth floor of a building which was on a hilltop (20m high).[3] In October 2004 the instruments were moved to the SMEAR III station about 200 m away from the previous site (Kumpula SMEAR III).[4] The surroundings of the measurement sites were very heterogeneous consisting of buildings, parking lots, roads, patchy forest and low vegetation. At a distance of 200 m from the SMEAR III station there was one of the major highways providing significant source of traffic emissions.

Rome

PNC has been continuously measured, starting from the framework of the European multi-center project “Health Effects of Air Pollution in Susceptible Subpopulations” (HEAPPS) (2001) until 2010, at the measurement station located in the front yard of the “Istituto Superiore di Sanità” (“ISS”) (2 km east of the city center). Inlets for particle measurements were at about 8 m from the curb and about 20 m from the street, Viale Regina Elena. Sampling height was 2 m from the ground. A flow of 25,000 vehicles/d was estimated. The area is not subject to industrial emissions. This site could be considered as a traffic-oriented site. PNC was measured by a TSI model 3022A condensation particle counter (CPC). The HEAPPS study QA/QC protocol has been followed.

Stockholm

PNC has been measure since May 2001 up until November 2005 at an urban background station located on Rosenlundsgatan. Then it was moved to Torkel Knutssonsgatan, ca 500 m to the north within the same area of the city. Parallell measurements have shown very similar levels at the two sites. Both sites are on a rooftop platform (25m height) and not directly affected by nearby emissions. Air quality dispersion modelling of number concentrations and NO_x has shown that Rosenlundsgatan and Torkel Knutssonsgatan represents well an urban background of the city of Stockholm.[5]

PNC has been measured simultaneously at the street and urban background site with CPC’s. Total number of particles with an aerodynamic diameter of 7 nm was measured using a CPC3022 (TSI Inc.), and a CPC3775 was used to measure total number of particles larger than 4 nm. Comparisons have shown that that many particles are between 4 nm and 7 nm (unpublished).

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Table S1. General information about the PNC measurements

City	Time period	Site ID	Site type[a]	Number of valid days	Number of missing days	Monitor, sampling method, etc.	Size range
Barcelona	13.7.05-31.1.09	Ba_IJA	UB	726	573 (44.1%)	WCPC 3785	5 nm - 1000 nm
Barcelona	9.2.09-31.12.10	BA_TG	UB	608	83 (12.0%)	WCPC 3785	5 nm - 1000 nm
Copenhagen	15.05.01–31.12.10	Co_HCOE	UB	1897	1621 (46.1%)	custom built DMPS using TSI CPC 3010	10 nm- 110 nm
Helsinki	06.03.01–07.10.04	Kumpula1	UB	1281	31 (2.4%)	DMA TSI307 + CPC TSI3022	10 nm- 100 nm
Helsinki	13.10.04–31.12.10	Kumpula SMEAR III	UB	2234	37 (1.6%)	Hauke-type DMA + CPC TSI3025 Hauke-type DMA + CPC TSI3010	10 nm- 100 nm
Rome	12.04.01–31.12.10	Rome_15	UT	2226	1325 (37.3%)	TSI 3022A	7 nm - 3 µm
Stockholm	04.05.01–31.07.05	Sto_Rosenlundsgatan	UB	1260	290 (18.7%)	TSI 3022	7 nm - 3 µm
Stockholm	17.02.08–31.12.10	Sto_Torkel	UB	893	156 (14.9%)	TSI 3775	4 nm - 3 µm

[a]UB=Urban Background; UT=Urban Traffic

Table S2. Specific characteristics of the PNC measurement stations

Site ID	Coordinates	Distance to nearest street (m)	Distance to nearest major street (m)	Distance to nearest intersection (m)	Street configuration	Sampling height (m)	Remarks
Ba_IJA	41°23'5.01"N 2° 7'9.00"E	50	150	70	Interrupted rows of homes on both sides of the road	10	Rooftop, large parking lot within 100 meter
Ba_TG	41°23'22.19"N 2° 6'59.43"E	50	280	60	Garden	2	
Co_HCOE	55°41'59.7"N 12°33'40.7"E	40	40	190	Interrupted rows of homes on both sides of the road	20	Rooftop
Kumpula1	60°12'17.2"N 24°57'50.1"E	10	120	20	Minor street on an university campus	18	4th floor of a building
Kumpula SMEAR III	60°12'10.4"N 24°57'40.0"E	100	160	220	On an university campus	4	parking lot within 50 m
Rome_15	41°54'16.9"N 12°31'02.9"E	20	20	165	Uninterrupted rows of homes on both sides of the street	2	
Sto_Rosenlundsgatan	59°18'40.6"N 18°3'28.7"E	20	60	80	Interrupted buildings on both sides of nearest street	20	Rooftop location
Sto_Torkel	59°18'57.7"N 18°03'28.0"E	20	240	20	Interrupted buildings on both sides. Park area on opposite side	24	Rooftop

Figure S1. Time-series plots of UFP measurements in the participating cities.

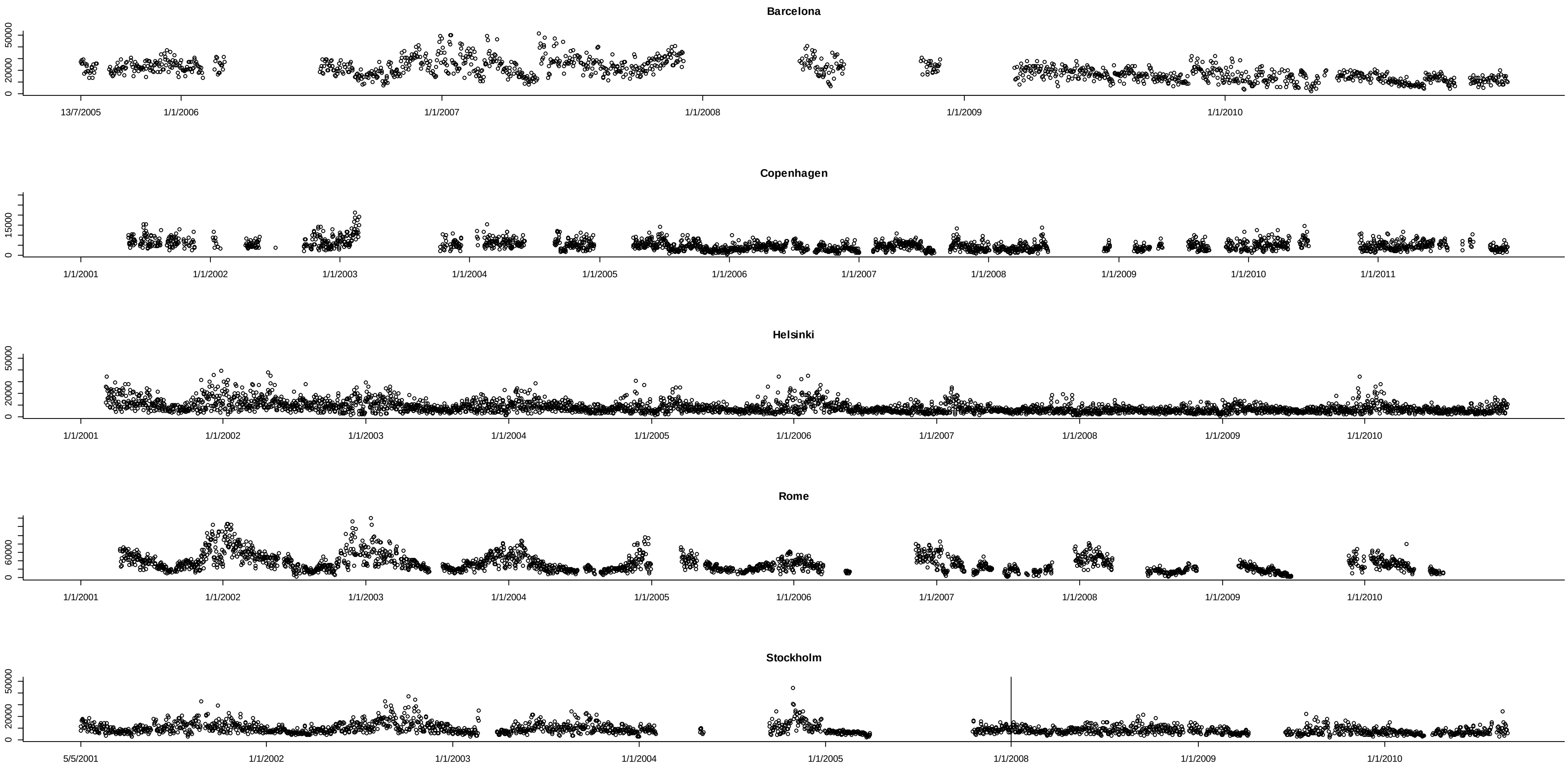


Figure S2. City-specific time-series plots of respiratory hospitalizations.

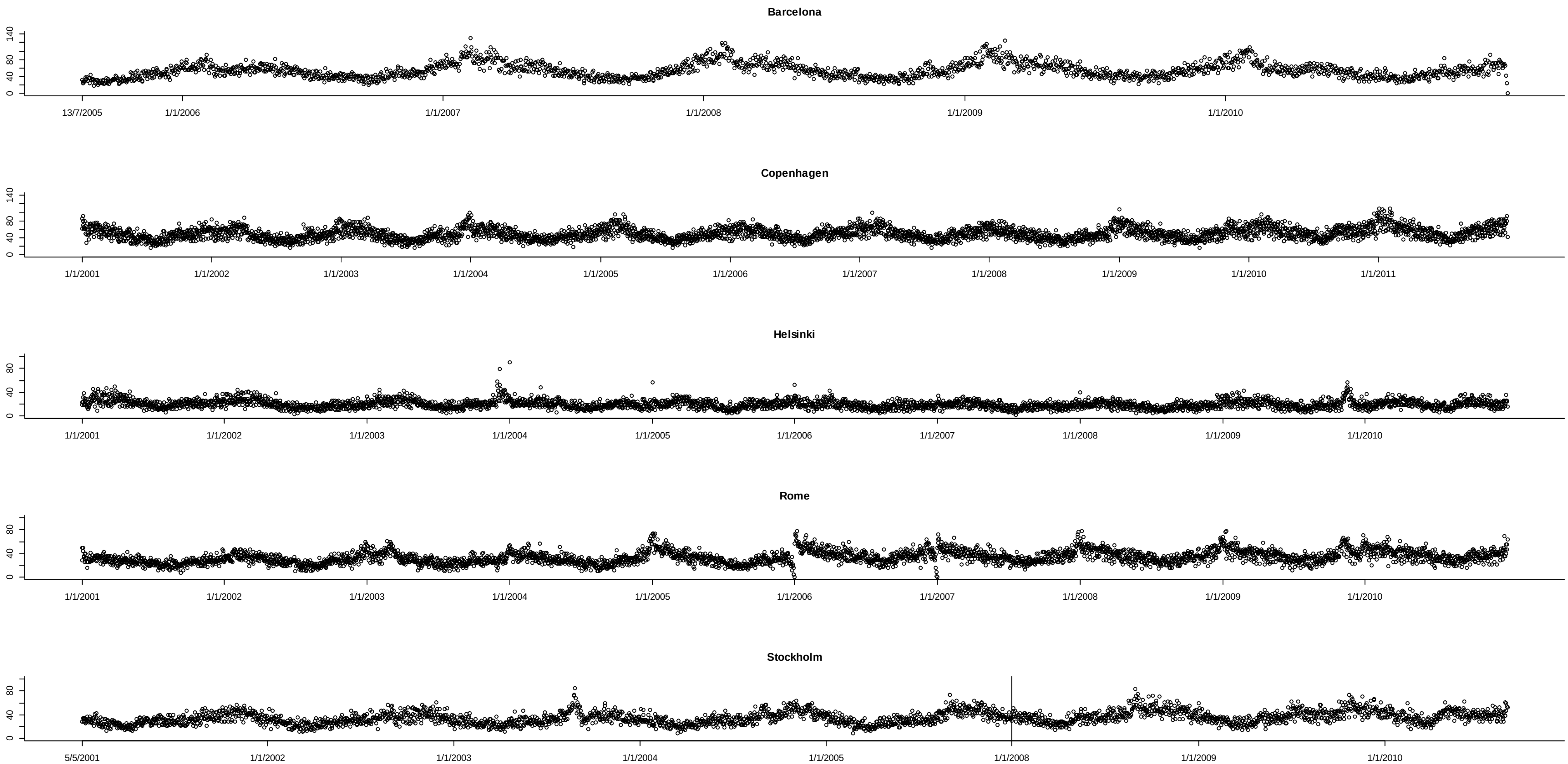


Table S3. Correlation coefficients between UFP number concentration per m^3 and NO_2 per city for the whole period and by warm/cool period.

City	Whole period	Warm period (April-September)	Cool period (October-March)
Barcelona	0.38	0.38	0.37
Copenhagen	0.43	0.46	0.54
Helsinki	0.69	0.60	0.74
Rome	0.64	0.51	0.59
Stockholm	0.64	0.56	0.62

Table S4. Pooled percent change (and 95% confidence intervals (CI)) in respiratory hospitalizations in five European cites per 10,000 increase in UFP number concentration per cm^3 and 10 μg/m^3 in PM_{10}, $PM_{2.5}$ and $PM_{2.5-10}$ for individual lags 0-10.

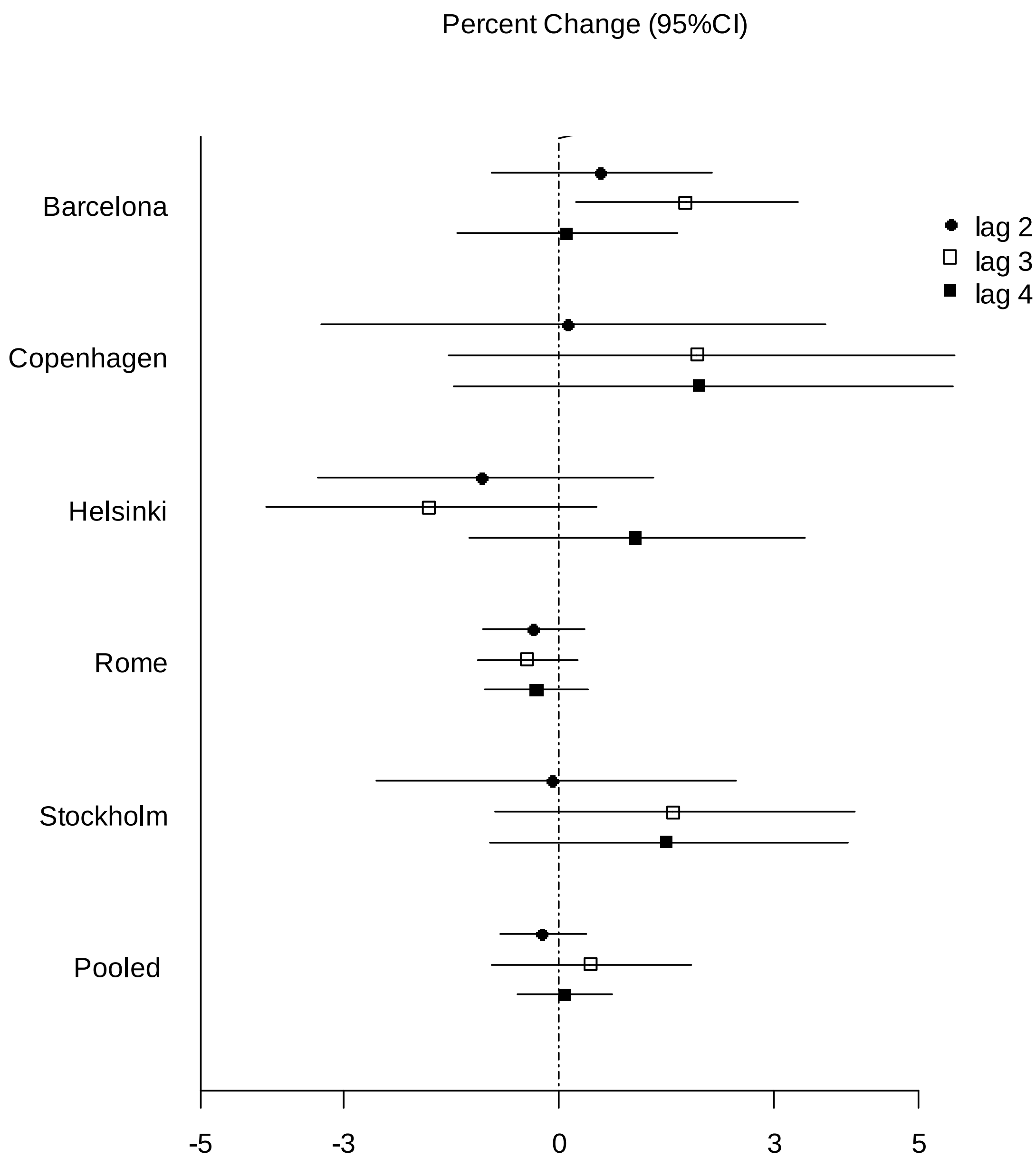
Lags	**UFP (number/cm^3)**	**PM_{10} (μg/m^3)**	**$PM_{2.5}$ (μg/m^3)**	**$PM_{2.5-10}$ (μg/m^3)**
0	-0.44 (-1.73, 0.87)*	**0.53 (0.17, 0.89)**	0.51 (-0.24, 1.27)*	**0.84 (0.14, 1.56)**
1	-0.58 (-1.93, 0.79)*	**0.67 (0.13, 1.22)***	**0.68 (0.01 ,1.36)**	0.45 (-0.46, 1.37)
2	-0.22 (-0.82, 0.38)	0.53 (-0.11, 1.18)*	0.57 (-0.37, 1.53)*	0.07 (-0.65, 0.79)
3	0.43 (-0.94, 1.83)*	**0.55 (0.05, 1.06)***	0.63 (-0.32, 1.60)*	0.48 (-0.23, 1.20)
4	0.07 (-0.59, 0.73)	**0.41 (0.06, 0.77)**	0.42 (-0.35, 1.20)*	0.38 (-0.34, 1.09)
5	0.43 (-0.58, 1.45)*	0.30 (-0.06, 0.65)	0.44 (-0.16, 1.04)	0.25 (-0.69, 1.20)
6	0.38 (-0.78, 1.56)*	0.23 (-0.27, 0.72)*	0.51 (-0.28, 1.32)*	-0.10 (-0.79, 0.61)
7	-0.37 (-1.39, 0.66)*	-0.13 (-0.56, 0.30)	0.01 (-0.95, 0.98)*	-0.37 (-1.06, 0.32)
8	-0.12 (-0.71, 0.47)	-0.22 (-0.64, 0.19)	0.01 (-0.83, 0.85)*	-0.62 (-1.32, 0.08)
9	-0.74 (-2.35, 0.89)*	-0.07 (-0.46, 0.31)	0.15 (-0.56, 0.87)*	-0.38 (-1.68, 0.93)*
10	0.39 (-0.20, 0.98)	-0.04 (-0.41, 0.33)	0.30 (-0.22, 0.82)	-0.26 (-1.76, 1.27)*

*Heterogeneity as defined by I^2>40% and p<0.20

In bold: statistically significant results.

Figure S3. City-specific and pooled percent change (and 95%confidence intervals (CI)) in respiratory hospitalizations for all ages in five European cites per 10,000 increase in UFP number concentration per cm^3 for lags 2 to 4, during the whole year (a) and during the warm period (April – September, (b)).

(a) Whole year



(b) Warm Period (April-September)

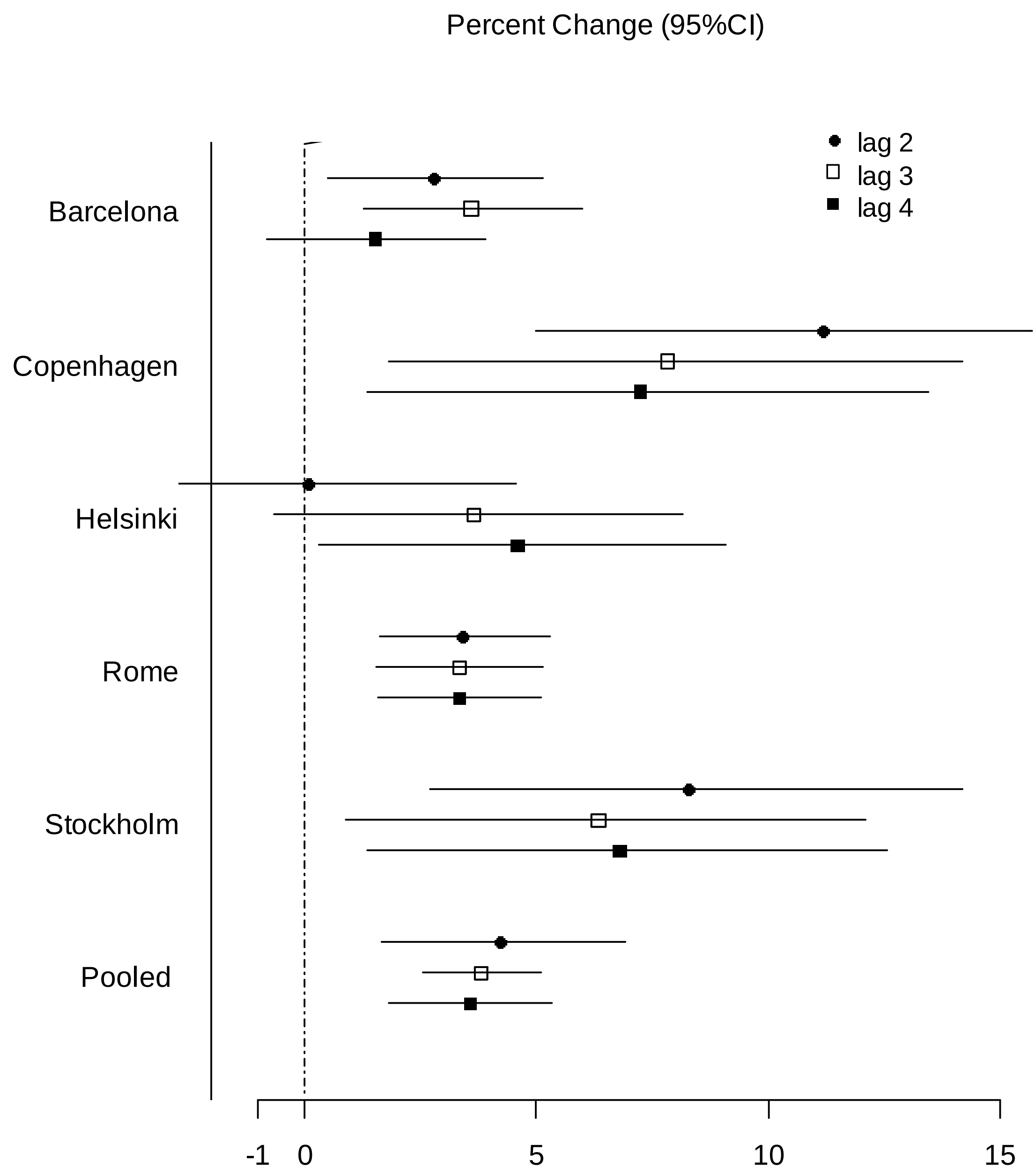


Table S5. Heterogeneity measures (I^2 (Q test p-value)) for the associations between UFP and respiratory admissions at different lags.

Lags	All ages	0-14 years old	15-64 years	65+ years	LRTI	COPD	Asthma
0	62% (0.031)	47% (0.110)	72% (0.007)	68% (0.014)	63% (0.030)	56% (0.058)	0% (0.729)
1	65% (0.023)	58% (0.051)	72% (0.006)	26% (0.245)	65% (0.023)	27% (0.244)	0% (0.516)
2	0% (0.784)	0% (0.877)	0% (0.596)	52% (0.081)	0% (0.404)	28% (0.236)	0% (0.715)
3	66% (0.020)	0% (0.668)	0% (0.794)	77% (0.002)	54% (0.070)	30% (0.220)	0% (0.648)
4	5% (0.377)	36% (0.183)	0% (0.743)	57% (0.055)	0% (0.529)	0% (0.686)	0% (0.425)
5	40% (0.154)	80% (0.001)	24% (0.264)	73% (0.005)	43% (0.137)	36% (0.180)	17%(0.309)
6	54% (0.070)	15% (0.318)	0% (0.985)	85% (<0.001)	5% (0.381)	73% (0.005)	3% (0.388)
7	44% (0.128)	0% (0.655)	12% (0.335)	60% (0.039)	49% (0.100)	0% (0.56)	53% (0.072)

LRTI: Lower Respiratory Tract Infections; COPD: Chronic Obstructive Pulmonary Disease.

Table S6. Pooled percent change (and 95% confidence intervals (CI)) in respiratory hospitalizations in five European cites, during the whole and the warm period, per10 μg/m^3 increase in NO_2 for individual lags 0-7. Results from single and two pollutants models.

Lags	**Whole period**		**Warm period**	
	NO_2 (μg/m^3)		**NO_2 (μg/m^3)**	
	Single pollutant	Controlling for UFP	Single pollutant	Controlling for UFP
0	-0.30 (-0.79, 0.19)	-0.14 (-0.84, 0.57)	1.69 (0.35, 3.06)	1.52 (0.37, 2.69)
1	-0.35 (-0.76, 0.07)	-0.17 (-0.66, 0.32)	2.02 (0.64, 3.41)	2.10 (0.70, 3.52)
2	-0.26 (-0.67, 0.15)	-0.37 (-0.86, 0.12)	1.83 (0.94, 2.73)	1.36 (0.48, 2.25)
3	0.43 (-0.40, 1.27)	0.54 (-0.39, 1.47)	2.40 (1.68, 3.11)	1.81 (1.10, 2.54)
4	0.59 (0.18, 1.01)	0.77 (0.27, 1.26)	2.31 (1.23, 3.41)	1.68 (0.78, 2.58)
5	0.97 (0.55, 1.39)	0.77 (0.27, 1.26)	2.61 (1.37, 3.87)	2.16 (1.37, 2.96)
6	0.82 (0.08, 1.57)	0.92 (0.10, 1.74)	2.20 (1.29, 3.12)	2.25 (0.89, 3.62)
7	0.32 (-0.25, 0.90)	0.54 (-0.10, 1.18)	1.17 (0.49, 1.84)	1.18 (0.37, 2.00)

Figure S4. Pooled percent change (and 95%confidence intervals (CI)) in respiratory hospitalizations for all ages in five European cites per 10,000 increase in UFP number concentration per cm^3 during the warm period of the year (April - September). Results from sensitivity analysis adjusting for NO_2 or CO or excluding Rome.