



# The influence of specific atmospheric circulation types on PM<sub>10</sub>-bound benzo(a)pyrene inhalation related lung cancer risk in Barcelona, Spain



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## ABSTRACT

Benzo(a)pyrene (BaP) is a Polycyclic Aromatic Hydrocarbon (PAH) well known for its carcinogenic effects. In this study, BaP levels in daily PM<sub>10</sub> samples collected at 8 stations in Barcelona (Spain), during the years 2013–2015, were analyzed in relation to distinct atmospheric circulation patterns. Our objective was to estimate the BaP inhalation related Lung Cancer Risk (LCR) in connection with the prevailing synoptic conditions. Air masses were also analyzed in order to examine the possibility of transboundary BaP contributions. The influence of high pressure anticyclonic systems caused a sharp increase of PM<sub>10</sub>-bound BaP concentrations in all stations due to recirculation and accumulation of polluted air, whilst the calculated BaP inhalation related LCR values implied a potential health risk from BaP exposure and were not recommendable primarily at central heavily trafficked sites. However the LCR remained below the upper limit posed by United States Environmental Protection Agency (US EPA), even under the most stagnant atmospheric conditions. The elaboration of backward air mass trajectories with Concentration Weighted Trajectory (CWT) algorithm indicated that combustion emissions in Spain, France and the industrialized Northern coast of Algeria are potential contributors to the PM<sub>10</sub>-bound BaP concentrations measured in Barcelona.

## 1. Introduction

Elevated concentrations of inhalable PM<sub>10</sub> (particles with an aerodynamic diameter smaller than 10 μm) are widely considered as a potential threat for human health. Particularly in the urban area of Barcelona, where our study is focused, Brines et al., 2016 discovered eight main aerosol sources affecting PM<sub>10</sub> concentrations: (1) vehicle exhaust and wear (10–27% of PM<sub>10</sub> mass on average), (2) road dust (8–12%), (3) mineral dust (13–26%), (4) aged marine (13–20%), (5) heavy oil (2%), (6) industrial (3–5%), (7) sulfate (11–17%) and (8) nitrate (17–21%). The estimated contribution of Barcelona's harbour emissions to the urban background reached 9–12% for PM<sub>10</sub> and 11–15% for PM<sub>2.5</sub> (particles with an aerodynamic diameter smaller than 2.5 μm) and is linked to primary emissions from fuel oil combustion but also to the formation of secondary aerosols (Pérez et al., 2016). The PM<sub>10</sub> diurnal variation at three urban sites in Barcelona was

influenced by traffic rush hour, whilst PM<sub>1</sub> (particles with an aerodynamic diameter smaller than 1 μm) concentrations at the Montseny regional background site (located within a regional natural park at about 50 km to the north–north-east side of the city of Barcelona) were clearly dependent on the sea breeze circulation bringing pollution from the city during the afternoon (Dall'Osto et al., 2013). During winter, maximum PM<sub>1</sub> levels in Barcelona were associated with strong thermal inversions favoring the accumulation of pollutants, whereas the lowest PM<sub>1</sub> concentrations were measured when the advection of air masses from the Atlantic Ocean caused the removal of the previously accumulated pollution (Jorba et al., 2013). During the years 2004–2014, statistically significant decreasing trends were observed for the contributions to PM (PM<sub>10</sub> and PM<sub>2.5</sub>) from industrial (metallurgy mainly) emissions in Barcelona, confirming the effectiveness of pollution control measures implemented at European or regional/local levels (Pandolfi et al., 2016). Additionally, an experiment carried out by

*Abbreviations:* PM, particle matter; PAHs, polycyclic aromatic hydrocarbons; EU, European Union; IARC, International Agency for Research on Cancer; BaP, benzo(a)pyrene; EEA, European Environment Agency; AOT, Aerosol Optical Thickness; AE, Angstrom Exponent; AERONET, Aerosol Robotic Network; SLP, Sea Level Pressure; ECAD, European Climate Assessment & Dataset; AGL, Above Ground Level; PLU, Plaça de la Universitat; ZUN, Zona Universitaria; POB, Poble Nou; SAN, Sants; EIX, Eixample; GSG, Gracia-Sant Gervasi; VDH, Vall D'Hebron; VER, Verdaguer; BAS, Barcelona Aeronet Station; BZU, Barcelona - Zona Universitaria; NCEP/NCAR, National Centers for Environmental Prediction/National Center for Atmospheric Research; NOAA, National Oceanic and Atmospheric Administration; US EPA, United States Environmental Protection Agency; LCR, Lung Cancer Risk; EC, exposure concentration; CA, average concentration; ET, exposure time; EF, exposure frequency; ED, exposure duration; AT, averaging time; IUR, Inhalation Unit Risk; WHO, World Health Organization; HYSPLIT, Hybrid Single-Particle Lagrangian Integrated Trajectory; CWT, Concentration Weighted Trajectory; NW, northwest; UK, United Kingdom; SW, southwest; NE, northeast; SE, southeast; ARL, Air Resources Laboratory

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Fig. 1. Map of Barcelona which includes the positions of a) background air pollution stations (blue dots) b) traffic air pollution stations (red dots) c) AERONET station (orange dot) and d) ECAD meteorological station (purple dot). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Amato et al., 2009, in one of the busiest roads of the city center of Barcelona, showed that a mean reduction of 4–5  $\mu\text{g}/\text{m}^3$  (7–10%) of kerbside  $\text{PM}_{10}$  concentrations was induced by street wash activities in the 24 h after the treatment.

Polycyclic Aromatic Hydrocarbons (PAHs) are chemical substances famous for their mutagenic, carcinogenic and teratogenic effects (Kim et al., 2013). In laboratory studies, animals have suffered from lung cancer after being exposed through inhalation to certain levels of some PAHs over long periods (Hecht, 1999, International Agency for Research on Cancer (IARC), 2012). Moreover, Zhang et al., 2016 reported that the cancer risk in the continental United States, due to inhalation exposure to the outdoor concentrations of several carcinogenic PAHs, was predicted to be significant. In the European Union (EU), ambient air legislation (Directive 2004/107/EC) aims at controlling levels of benzo(a)pyrene (BaP) contained in  $\text{PM}_{10}$  (annual target level 1  $\text{ng}/\text{m}^3$ ) since this compound carries the highest toxic load of any airborne PAH (Barrado et al., 2013). Various studies which examine  $\text{PM}$ -bound PAH levels have been conducted in Barcelona. Van Drooge and Grimalt, 2015, conducted sampling campaigns at an urban and a rural site in the broader area of Barcelona to collect atmospheric particles of different sizes ( $> 7.2$ ;  $7.2$ – $3$ ;  $3$ – $1.5$ ;  $1.5$ – $1$ ;  $1$ – $0.5$ ;  $< 0.5$   $\mu\text{m}$ ). Overall,  $> 70\%$  of the sum of all quantified PAHs were present in the fraction  $< 0.5$   $\mu\text{m}$ , whilst around 10% was present in particles  $> 3$   $\mu\text{m}$ . The dominance of PAHs in the smallest fraction marked the combustion processes as a major source of these particles. Reche et al., 2012, also concluded that PAHs in Barcelona are predominantly present in the fine particles ( $\text{PM}_1$ ) and that there is no significant contribution from coarse particles ( $\text{PM}_{10}$ – $\text{PM}_{2.5}$ ) to these compounds. The total concentrations of 12 PAHs, determined by Alier et al., 2013 in  $\text{PM}_1$  samples collected at an urban background site and a road site in Barcelona, were two times higher at the road site (1.91  $\text{ng}/\text{m}^3$ ) than at the

urban background site (0.89  $\text{ng}/\text{m}^3$ ), reflecting a stronger influence of residues from fossil fuel vehicles at the former site. Temperature inversions increased the levels of  $\text{PM}_1$ -bound PAHs in the urban domain of Barcelona due to atmospheric stagnation, whereas strong sea breeze was associated with transport of  $\text{PM}_1$ -bound PAHs from the polluted coastal areas towards inland regions (Van Drooge et al., 2012).

Previous publications have estimated the cancer risk related to lifetime inhalation exposure at the ambient PAH concentrations measured in several Spanish conurbations. In the study of de la Gala Morales et al., 2015, an annual  $\text{PM}_{10}$ -bound BaP average concentration of 0.09  $\text{ng}/\text{m}^3$  was determined in Extremadura, implying a lifetime lung cancer risk of  $7.83 \times 10^{-6}$  (approximately eight inhabitants per million people may develop lung cancer due to the exposition to BaP in atmospheric particulates). Callén et al., 2013 reported that peak lifetime lung cancer risk values in Zaragoza were associated with higher contribution of stationary and vehicular emissions in winter season and were also favored by high relative humidity, low temperature and low wind speed. In this study, BaP inhalation related lung cancer risk in trafficked and urban background areas of Barcelona was estimated in connection with atmospheric circulation patterns. Air masses were also analyzed in order to examine the possibility of exogenous BaP contributions.

## 2. Data and methodology

### 2.1. Data

The air quality database (AirBase) of the European Environment Agency (EEA) of the EU (<https://www.eea.europa.eu>), contains air quality monitoring data and information submitted by participating countries throughout Europe. A new dataset of ambient concentrations

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