AIRUSE-LIFE+: 2003-2014 trends of PM_{10} and PM_1 source contributions in Barcelona, NE Spain.

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In the last decades, the European Union has made large efforts for the improvement of air quality (AQ) by means of the elaboration and implementation of both ambient air and emissions directives. Additional measures to abate pollution have been applied by various Member States, as well as by regional and city governments.

Among the efficiently implemented standards in emissions abatement are the IPPC (Integrated Prevention and Pollution Control) Directive (1996/61/EC, Industrial Emissions Directive 2010/75/EC), the Large Combustion Plants Directive (2001/80/EC), the National Emission Ceilings (NEC) Directive (2001/81/EC), the EURO standards on road traffic emission (1998/69/EC, 2002/80/EC, 2007/715/EC). Furthermore, IMO (International Maritime Organization)/MARPOL and the EU have set absolute limits on sulphur content in fuel, and SOx and NOx emissions from ships (Directive 2005/33/EC). Thus, there is a clear evidence that PM concentrations have decreased markedly during the last decade in a number of European regions (EEA, 2013), as a result of: a) the EU policy for reducing emissions; b) the national and numerous regional and local AQ plans implemented, and c) the favourable 2008-2012 meteorology for Southern Europe as compared with 2005-2007.

This study seeks to evaluate the sources of anthropogenic air pollutants in the Barcelona Metropolitan Area (BMA; Spain), with special emphasis on the study of the trends of the source contributions over the period 2003-2014. Source apportionment was performed by means of ME-2 model, a flexible program that permits the incorporation of any a priori information such as chemical properties or linear constraints into the model as a target to be fit to some specified precision (Amato et al., 2009). The observed trends were related with the major EU/regional/local pollutants abatement strategies in order to evaluate the effectiveness of these strategies.

Seven pollutants sources were identified in both PM_{10} and PM_1 fractions as *Vehicle exhaust*, *Road dust*, *Secondary nitrate*, *Secondary sulphate*, *Mineral* (including the Saharan dust contribution in PM_{10}), *Heavy oil combustion* and *Metallurgy* processes. An eighth factor detected in PM_{10} was an *Aged sea salt factor*, not detected in PM_1 given that this source mainly contributes to the coarse PM mode (PM_{1-10}). Temporal trends were analysed by means of the Theil-Sen method available in the Openair software (Carslaw and Ropkins, 2012).

All detected source contributions in Barcelona showed statistically significant decreasing trends with the exception of the marine contribution to PM_{10}.

As an example, Figure1 shows the contributions of *Secondary sulphate* and *Secondary nitrate* which clearly decreased with time in both fractions. These decreases were related to the implementation of the EC Directive on Large Combustion Plants and the ban of the 2007 Regional AQ Plan on the use of heavy oils and petroleum coke for power generation and to lower NO_x emissions from power generation plants due to the economic crisis, the implementation of a large combustion plants directive, and the modifications in the Barcelona public transport (SCRT, hybridization and shift to natural gas engines).

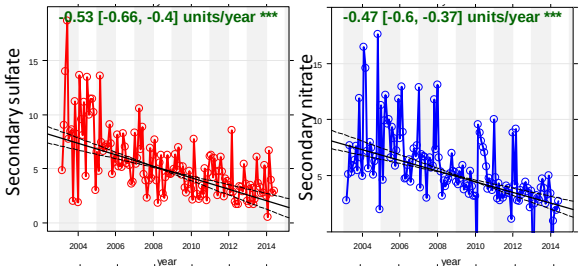

Figure 1. Temporal trends of *Secondary sulphate* and *Secondary nitrates* PM_{10} source contributions from ME-2 analysis. P-value: degree of statistical significance. ***: $p<0.001$. Variations expressed as mass (μgm^{-3}) per year, including minimum and maximum ranges.

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