Trends of PM source contributions and chemical tracers in NE Spain during 2004 - 2014: A multi-exponential approach

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Meeting the air quality (AQ) standards is one of the major environmental objectives to protect people from breathing air with high levels of pollution. Many studies have been published in these last years showing clearly that the concentrations of particulate matter (PM), and other air pollutants such as sulphur dioxide (SO_2) and carbon monoxide (CO), have markedly decreased during the last 15 years in many European Countries (i.e. EEA, 2013; Querol et al., 2014)

In this work data from two twin stations (Barcelona (BCN), urban background, and Montseny (MSY), regional background; i.e. Pandolfi et al., 2014), located in NE of Spain, were used to study the trends of the concentrations of different chemical species in PM_{10} and $PM_{2.5}$ along with the trends of the PM_{10} source contributions from Positive Matrix Factorization (PMF) model. Eleven years of chemical data (2004–2014) were used for this study. Trends of both specie concentrations and source contributions were studied using the Mann-Kendall test for linear trends and a new approach based on multi-exponential fit of the data (i.e. Figure 1). Despite the fact that different PM fractions (PM_1, $PM_{2.5}$, PM_{10}) showed linear decreasing trends at both stations, the contributions of specific sources of pollutants and the related chemical tracers showed exponential (single or double) decreasing trends. The different types of trends observed reflected the different effectiveness and/or time of implementation of the measures taken to reduce the concentrations of atmospheric pollutants (i.e. those implemented in the Industrial Emission Directives and in the Large Combustion Plants Directive). Moreover, the trends of the contributions from specific sources such as those related with industrial activities and with primary energy consumption mirrored the effect of the financial crisis in Spain from 2008. The sources that showed statistically significant downward trends at both BCN and MSY during 2004-2014 were *Ammonium sulfate*, *Ammonium nitrate*, and *V-Ni bearing* source. The contributions from these sources decreased exponentially during the considered period indicating that the observed decrease was not gradual and consistent over time. Moreover, statistically significant decreasing trends were observed for the contributions to PM from the *Anthropogenic* source at MSY (mixed metallurgy and road traffic) and from the *Industrial* (metallurgy mainly) source at BCN. These sources were clearly linked with anthropogenic activities and the observed decreasing trends confirmed the effectiveness of pollution control measures implemented at EU or regional/local levels. The general trends observed for the calculated PMF source contributions well reflected the trends observed for the chemical tracers of these pollutant sources.

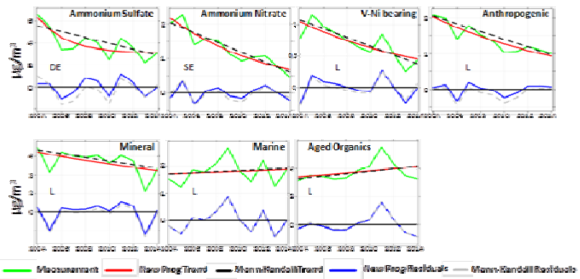

Figure 2. Mann-Kendall and Multi-exponential trends for source contributions in PM_{10} at MSY. Measured concentration (green line); Multi-exponential trend (red line); Multi-exponential residuals (blue line); Mann-Kendall trend (black line); Mann-Kendall residuals (grey line). Trend type: linear (L), single-exponential (SE), double exponential (DE).

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