Trends of PM2.5 and Chemical Composition in at a Regional Backround Site in the Western Mediteranean over the Past Nine Years

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TRENDS OF $PM_{2.5}$ AND CHEMICAL COMPOSITION IN AT A REGIONAL BACKGROUND SITE IN THE WESTERN MEDITERANEAN OVER THE PAST NINE YEARS

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INTRODUCTION

The directive 2008/50/EC establishes limit and target values (annual average of 25 µg m^{-3}) for ambient air $PM_{2.5}$ in all member states of the European Union (EU). Although the target values for $PM_{2.5}$ did not come into force until 2010 (with limit values being enforced in 2015), levels of $PM_{2.5}$ have been decreasing throughout Europe for a number of years, as outlined in this paper. Thus, it can be reasonably assumed that this Europe-wide reduction is a result of the implementation of emission abatement strategies enforced within the EU and the introduction of the Integrated Pollution Prevention and Control (IPPC) directive. Other factors appear to have had some influence on PM concentrations observed over the past decade. Since mid-2008, Europe and much of the developed world has been in the grips of a severe economic recession which, at the time of writing this article, appears to show no sign of abating. Indeed, this economic crisis has been most severely felt in the peripheral economic states of Europe such as Spain and Portugal, and Europe's fourth largest economy, Italy. Anthropogenic activities have long been associated with air pollution, through fuel oil combustion, industrial activities, traffic emissions and construction, to name a few. The economic recession has severely impacted these activities, and the possible resulting effect is a net reduction in pollution. Furthermore, large scale meteorology can also influence regional ambient concentrations of aerosols, such as the influence of the North Atlantic Oscillation. The $PM_{2.5}$ trends for many stations across Europe, and to a greater extent Spain, are analysed and compared to those of MSY. These trends are analysed for statistical significance, in order to determine if the decreases observed are gradual and uniform. Special focus is then given to the in-depth investigation into the temporal trends observed, not only for $PM_{2.5}$, but also to the various chemical components of $PM_{2.5}$ at MSY measured between 2002 and 2010.

METHODS

The Montseny station (MSY) is located in the Montseny natural park 40km to the NNE of the Barcelona urban area, and 25 km from the Mediterranean coast. The station is located on the upper walls of a valley extending perpendicularly from the Catalan Pre-Coastal ranges, in a densely forested area known as La Castanya. The station is situated relatively far from urban and industrial zones, but the region is generally densely populated and heavily industrialised, and local anthropogenic emissions can affect this site under specific meteorological conditions. Samples of $PM_{2.5}$ were collected on quartz fibre filters (Schleicher and Schuell, QF20 until 2009, Munktell thereafter) for 24 hour periods roughly once a week until 2007, and consecutively every four days from 2008, with high volume samplers (30 m^3/h) DIGITEL-DH80 and MCV-CAV, equipped with a $PM_{2.5}$ cut off inlet (manufactured by DIGITEL). Filters were analysed using different instrumental techniques to determine concentrations of a range of elements and components.

Data from various regional background stations across Europe were obtained from internet databases or through personal correspondence with the responsible bodies for the station. Temporal trend analysis was performed for all the stations where sufficient data was available by means of the nonparametric Mann-Kendall test for the trend and the nonparametric Sen's method for the magnitude of the trend.

RESULTS

Mean $PM_{2.5}$ levels recorded at MSY (determined gravimetrically) from 2002 to 2010 were 12.6 μg m^{-3}. $PM_{2.5}$ levels were elevated when compared with Spanish EMEP stations. The average $PM_{2.5}$ concentration for 10 EMEP RB sites across the Iberian Peninsula (IP) for the same time period was 8.6 μg m^{-3}, and the average value for two other RB stations in the NE IP (namely Els Torms and Cabo de Creus) was 10.5 μg m^{-3}. Thus, the MSY station registered higher levels of PM compared to average concentrations across Spain (+37%) and those registered from stations in the NE of Spain (+18%). This surplus may be attributed to anthropogenic influences. The greater area surrounding MSY, especially the valleys in the pre-coastal depression, is densely populated and highly industrialised, being a significant source of pollution reaching the MSY site. Comparing these values with other RB sites across Europe, concentrations measured at MSY were considerably higher (+34%) than those in Portugal (9.4 μg m^{-3}), Germany (10 μg m^{-3}) and Scandinavia (6.6 μg m^{-3}). They were slightly lower than levels recorded in Switzerland (14.5 μg m^{-3}) and significantly lower than levels recorded in Austria (19.7 μg m^{-3}). Ispra recorded the highest levels of all the stations included in this work, with $PM_{2.5}$ of 26.2 μg m^{-3}. Climate conditions are likely to influence $PM_{2.5}$ levels at each of the RB stations mentioned, whereby differences in precipitation levels and prevailing wind systems could account for the differing concentrations observed, especially for Atlantic and Scandinavian countries. Anthropogenic influences are likely to be most prevalent in Ispra in Northern Italy, as it is subjected to intense episodes of pollution owing to thermal inversions in winter and emissions from nearby heavy industry in the Po valley. High pressure weather systems over Eastern Europe in winter can lead to stagnant conditions across Austria and Switzerland, causing the accumulation of pollutants. Furthermore, the regions can be affected by long range transport of pollution from central and Eastern Europe, and biomass burning emissions in winter.

All the stations chosen for the trend analysis underwent decreases in ambient $PM_{2.5}$ concentrations to varying degrees. MSY recorded a drop of 35 % in ambient $PM_{2.5}$ concentrations over the measurement period of 2002-2010. This trend was followed more or less all across Europe. On average, a reduction of 32% has been observed in $PM_{2.5}$ levels since 2002 throughout Spain, 31% at Illmitz, 36% at Payerne, 34% at Ispra, 35% in Sweden, 32% across Germany, 32% in Finland, 41% in Norway and 38% in Portugal. What was most striking of these statistics were the similarities in reductions shown across Europe, indicating that the forces responsible must be similar across the continent. It can be reasonably assumed that this continuous and, in most cases, gradual reduction is a reflection on the efficacy of pollution abatement strategies employed by member states of the E.U. The E.U. Directive 2008/50/EC specifically controls ambient concentrations of PM_{10} and $PM_{2.5}$ and targets industrial emissions (IPPC Directive, 2008/1/EC), and these measures clearly had a direct effect on pollutant levels in Europe. As mentioned previously, all the stations included underwent a steady and gradual decrease for most of the duration of the study.

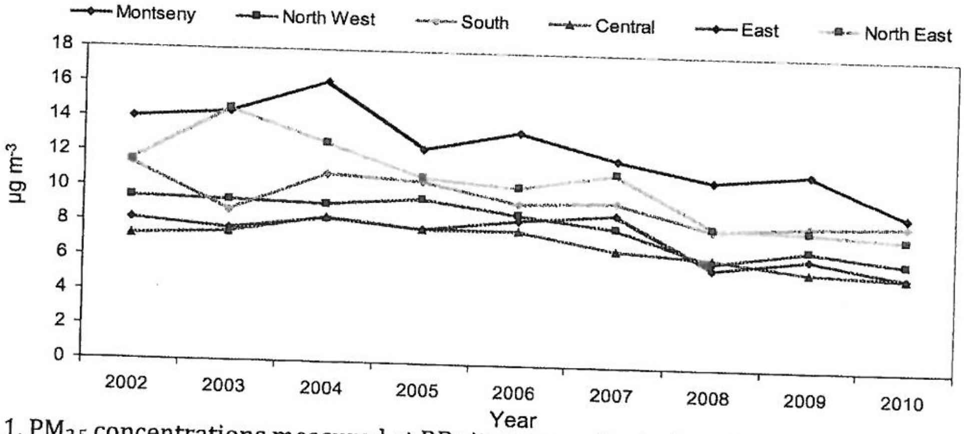

Figure 1. $PM_{2.5}$ concentrations measured at RB sites across Spain from 2002-2010.

As illustrated in Fig. 1, a marked decrease was observed for each area of Spain through the decade, with minimum concentrations occurring from 2008 onwards. Stations are categorised according to their location in Spain and values are mean $PM_{2.5}$ concentrations of the stations for that location for each year: Niembro and O Saviñao are categorised as North; Viznar and Barcarrota are South; Peñausende, Campisábolos and Risco Llano are considered Central; Zarra is East; Cabo de Creus and Els Torms are North East. For many regions levels reached a minimum in 2008, followed by a slight increase in 2009 (for all except central Spain), and a reduction once again in 2010. MSY followed a similar trend to that observed for the other stations in the NE peninsula, albeit with slightly higher levels measured in comparison. The fact that the PM levels here follow a similar trend verifies that the trend is real and observed across the region. In fact, this decreasing trend is observed for many stations across Europe (Fig. 2).

For some stations in Germany, Austria, Switzerland, Finland and Sweden, levels of $PM_{2.5}$ experienced noticeable decreases from 2007 onwards. For example, in Vavihill, Sweden and Waldhof in Germany, a sharp decrease was recorded of 4 $\mu g\ m^{-3}$ and 5 $\mu g\ m^{-3}$ between 2006 and 2007, respectively (Fig. 2). A similar reduction, although not quite as pronounced, was observed for stations Illmitz, Payerne, Utö, Aspverten, Virolahti, Schwartenberg and Schauinsland. This gradual reduction observed across Europe is possibly a direct result of the implementation of the aforementioned pollution abatement strategies. Indeed, the countries in which these stations are located have to a large extent avoided economic recession compared to the peripheral European states such as Spain, Portugal and Italy. Thus, reductions there have not been quite as pronounced as those recorded for the last two years in the IP and Ispra. However, it should be highlighted that PM levels in many of the stations, especially in Norway (mean $PM_{2.5}$ of 4.3 $\mu g\ m^{-3}$) and Finland (6.0 $\mu g\ m^{-3}$) for example, were comparatively low even at the beginning of the measurement period. Thus, even though a decreasing trend has been observed in these regions, it cannot be decisively ascertained that the economic recession and pollution abatement strategies have not impacted PM levels here, considering the low initial concentrations.

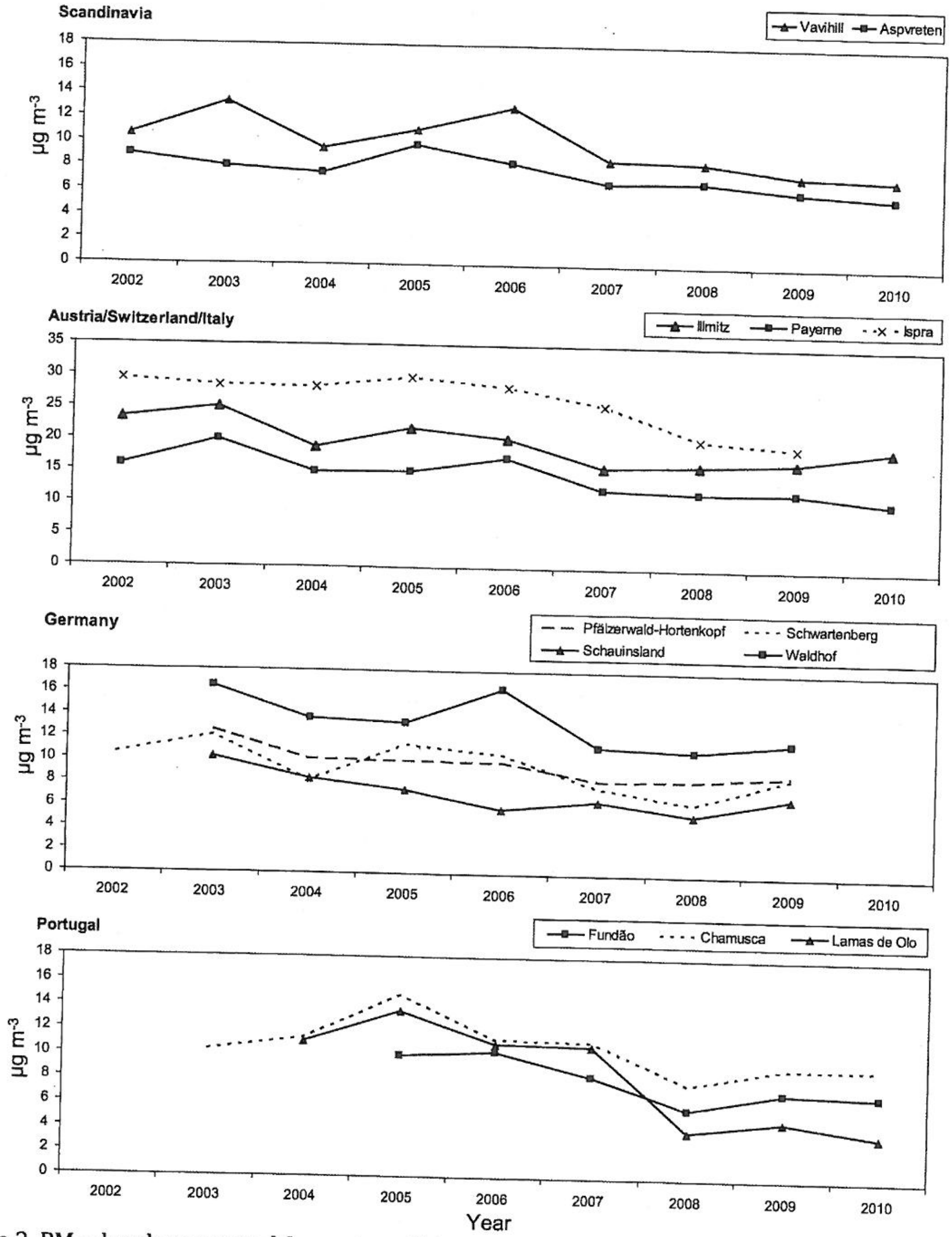

Figure 2. $PM_{2.5}$ levels measured for various RB stations across Europe according to AIRBASE and EMEP data.

The North Atlantic Oscillation (NAO) is a large scale oscillation in atmospheric mass, which is believed to be one of the most influential climate modes especially in winter in the North Atlantic region, influencing temperature, precipitation and wind speed across the European continent. The NAO index (NAOi) is a means of quantifying the fluctuations in the dominant pressure systems (the Azores high pressure system and the Icelandic low) over the Atlantic. When the NAO index is in a positive phase, strong winds, precipitation and mild temperatures from the Atlantic move across Northern Europe, and warm, dry weather is experienced in Southern Europe. However, when the NAO index is negative, the Atlantic weather fronts are directed toward a more southerly trajectory giving rise to wetter, windier weather across the Iberian Peninsula and colder weather across Northern Europe. During the warm winter of 2007, the NAOi was in a distinctly positive phase, whereas during winter 08/09 and winter 09/10, the NAOi was in a negative phase. These opposing phases of the NAO might have different implications for different areas of Europe. Whereas warmer conditions in winter for Northern Europe might equate to less domestic heating emissions, the same NAO conditions in Spain give rise to more frequent and more intense winter pollution episodes and stagnation of air masses. However, under negative phases of NAO the impact of these pollution episodes is diminished through dispersion and higher precipitation associated with Atlantic weather fronts. Indeed, winter 2010 was notable for two reasons; unusually high precipitation over the IP which coincided with one of the most negative NAOi since measurements began (Vincente-Serrano et al. (2011), and one of the coldest winters in decades across northern Europe. Incidentally, winter 2010 also registered the lowest PM levels recorded at MSY since 2002. Thus, the extreme negative phase of the NAO possibly had two important effects on ambient PM levels that winter at MSY: increased Atlantic advection and precipitation, and possibly less Saharan dust intrusions. As mentioned previously, the occurrence of Saharan dust intrusions over the IP can affect PM levels significantly. A linear relationship was observed between the frequency of NAF episodes (in days) and the corresponding NAO index for the winter months across the Iberian Peninsula (Fig. 3). This indicated that when NAO is more intensely positive, the probability of air masses from North Africa reaching the Iberian Peninsula is much higher. Conversely, when NAO is negative, intense Atlantic Advection directed over the Peninsula can block North African air masses and prevent these air masses moving northward.

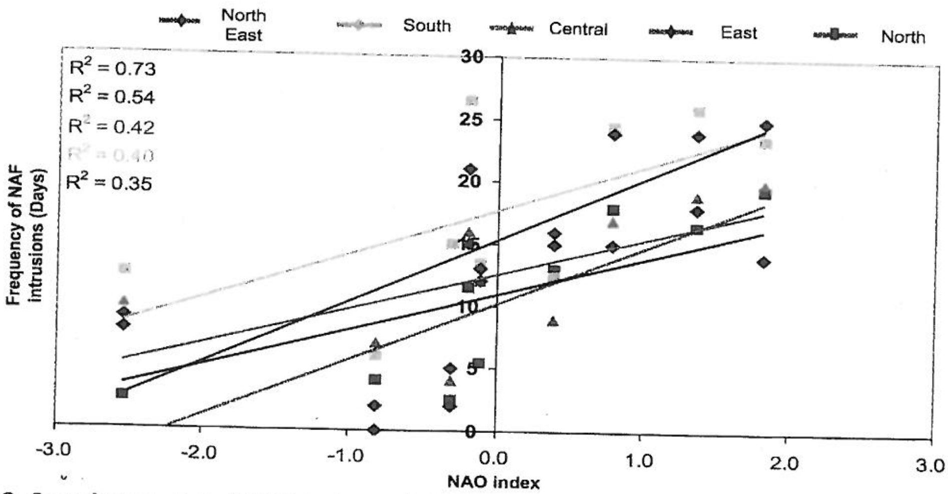

Figure 3. Correlation plot of NAO index and the frequency of Saharan dust intrusions (NAF) in days during winter for 2002-2010 for different regions of Spain.

Finally, one further influential factor may explain the decline in $PM_{2.5}$ concentrations observed. As outlined above, a declining trend was discernible throughout Europe, and variations around this trend are likely a result of meteorology. However, in some cases the annual variation in concentrations appeared to fluctuate less intensely relative to previous years. Furthermore, this appeared to be preceded by a drop in concentrations, specifically between 2007 and 2008. $PM_{2.5}$ levels thereafter fluctuate much less and in most cases do not exceed the linear decreasing trend. What the countries have in common where this occurred, such as in Spain, Portugal and Italy, is the on-going economic crisis. The countries which have been largely unaffected by the economic recession, such as Germany, Austria, Switzerland, Finland, Sweden and Norway, did not display any discernible larger decrease other than that which was observed for the previous years. For example, $PM_{2.5}$ concentrations at Ispra in the heavily industrialised Po valley in Northern Italy dropped 6 $\mu g\ m^{-3}$ between 2007 and 2008, and this decrease was maintained for the year after, removing the likelihood that the decrease was anomalous. An even more dramatic decrease was observed across Portugal, especially for one station (Lamas de Olo). At MSY, reductions in concentrations of typically anthropogenic aerosols sulphate, nitrate and organic carbon accounted for the majority of the reduction observed in $PM_{2.5}$ concentrations.

CONCLUSIONS

The findings in this article provide good evidence that the implementation of pollution abatement strategies in Europe is having a direct effect on the levels of $PM_{2.5}$ and its various components. It is also hypothesised that the current economic climate, in recession since mid 2008 in Spain and many countries in Europe, is also affecting atmospheric pollutants through a reduction in activities associated with a healthy economy (increased road traffic, industrial processes, construction etc.). A reduction in $PM_{2.5}$ concentrations has been observed in Spain and across Europe, and, in most cases, this reduction has been gradual and consistent over time, implying the success of cleaner anthropogenic activities. Additional to this progressive trend, in some cases and especially for RB stations in the IP and Ispra in northern Italy, a marked decrease has been recorded since 2008, coinciding with the beginning of the economic crisis. Finally, large scale meteorology has been shown to be an important influential factor on ambient aerosol concentrations across the Iberian Peninsula, with the North Atlantic Oscillation controlling the frequency of Saharan dust intrusions across the Iberian Peninsula in winter. Specifically at MSY, organic matter and secondary inorganic aerosol accounted for the majority of the reduction observed.

ACKNOWLEDGEMENTS

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