



Analysing the health effects of simultaneous exposure to physical and chemical properties of airborne particles



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ABSTRACT

Background: Airborne particles are a complex mix of organic and inorganic compounds, with a range of physical and chemical properties. Estimation of how simultaneous exposure to air particles affects the risk of adverse health response represents a challenge for scientific research and air quality management. In this paper, we present a Bayesian approach that can tackle this problem within the framework of time series analysis.

Methods: We used Dirichlet process mixture models to cluster time points with similar multipollutant and response profiles, while adjusting for seasonal cycles, trends and temporal components. Inference was carried out via Markov Chain Monte Carlo methods. We illustrated our approach using daily data of a range of particle metrics and respiratory mortality for London (UK) 2002–2005. To better quantify the average health impact of these particles, we measured the same set of metrics in 2012, and we computed and compared the posterior predictive distributions of mortality under the exposure scenario in 2012 vs 2005.

Results: The model resulted in a partition of the days into three clusters. We found a relative risk of 1.02 (95% credible intervals (CI): 1.00, 1.04) for respiratory mortality associated with days characterised by high posterior estimates of non-primary particles, especially nitrate and sulphate. We found a consistent reduction in the airborne particles in 2012 vs 2005 and the analysis of the posterior predictive distributions of respiratory mortality suggested an average annual decrease of –3.5% (95% CI: –0.12%, –5.74%).

Conclusions: We proposed an effective approach that enabled the better understanding of hidden structures in multipollutant health effects within time series analysis. It allowed the identification of exposure metrics associated with respiratory mortality and provided a tool to assess the changes in health effects from various policies to control the ambient particle matter mixtures.

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1. Introduction

Airborne particle matter (PM) is one of the air pollutants of primary health concern. Over the past two decades, PM mass metrics (e.g., particles with aerodynamic diameter $<10\ \mu\text{m}$, PM_{10} , and particles with aerodynamic diameter $<2.5\ \mu\text{m}$, $\text{PM}_{2.5}$) have received much attention, and many studies have shown that high concentrations of PM are associated with increased risks of mortality and morbidity. More recently, the evidence derived from studies of long- and short-term

exposure has been judged sufficient to infer causality for fine particles (EPA, 2012; WHO/Europe, 2013).

The evidence for the association between PM and short-term adverse endpoints, derives largely from observational ecological time series studies (e.g., Bell et al., 2004; HEI, 2010; Bell et al., 2013; Atkinson et al., 2014; and references therein). Since the early 1990s the results from these studies have played an important role in setting standards for acceptable levels of ambient pollution. The quantification of the impact of air pollution on health has been historically undertaken through a single pollutant approach, using regression-based techniques, where the co-pollutants have been treated as modifying or confounding factors. This reliance on single pollutant results is due, in part, to measurement and source complexities (such as the intrinsic correlated nature of air pollutants) which have limited the development of statistically robust multipollutant models, and in part to the regulatory strategies of

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air quality management which have addressed a single pollutant at a time (Dominici et al., 2010).

Air pollution exists, however, as a heterogeneous mix of different compounds. In particular, airborne particulate is made up of a number of solid and liquid components, including acids (such as nitrates and sulphates), organic chemicals, metals, soil or dust particles, soot, allergens and smoke. These components also vary in number, size, shape, surface area, solubility and origin. Thus, estimation of how simultaneous exposure to multiple air pollutants affects the risk of adverse health response represents a challenge for scientific research and air quality management.

To gain better insight into the features of air pollution mixtures and their effect, there is a consequent need to explore new statistical methods able to integrate standard methodological tools for a better understanding of these complex systems. In a recent review of techniques for estimating health effects of multiple air pollutants, Oakes et al. (2014) highlighted that clustering of pollution profiles has been shown to be an effective approach.

Previous temporal clustering analyses have been successfully applied in air pollution exposure assessment, involving mainly heuristic methods such as agglomerative hierarchical clustering (Gu et al., 2012) and *k*-means partitioning clustering (Austin et al., 2012). Recently, *k*-means clustering solutions of air pollutants have also been used as covariates within health model effect estimation (Matyasovszky et al., 2011; Zanobetti et al., 2014).

Despite the increasing popularity of these methods, they have some well known drawbacks. First, they do not allow an assessment of the statistical properties of the solutions provided, for example they do not provide an assessment of clustering uncertainties. Moreover, because these methods are based on similarity/dissimilarity measures between objects that are essentially described in terms of distance (e.g., Euclidean distance), they require that the time series of each pollutant has exactly the same dimensionality (i.e., they do not allow the inclusion of records which have missing data). This can represent a limitation when applied to air pollution monitoring data.

Mixture models (McLachlan and Peel, 2000; McLachlan and Baek, 2010) have been proposed as an alternative to heuristic clustering techniques. Generally, model-based clustering methods are based on the idea that the data follow a finite mixture of probability distributions such that each component distribution represents a cluster. Frühwirth-Schnatter and Kaufmann (2008) showed that model-based clustering based on finite mixture models can be extended to time series in a quite natural way. In the air quality field, Gómez-Losada et al. (2014) applied a finite mixture model for characterising air pollution mixtures, using maximum likelihood, via the expectation-maximization algorithm.

A long-standing issue that finite mixture models share with many traditional clustering methods (e.g., *k*-means), is the a priori determination of the number of clusters. Different methods can be used to estimate the number of components (i.e., clusters), using for example model selection criteria. However, an alternative way to handle this problem is to adopt a Bayesian nonparametric modelling approach, where the number of mixture components is not fixed in advance, but is determined by the model and the data. These models can be implemented using a Dirichlet process (DP) (Ferguson, 1973; Antoniak, 1974), a stochastic process commonly used in Bayesian nonparametrics to model the uncertainty about the functional form of the distribution of the parameters in a model. The support of the DP is restricted to discrete distributions and this results in a clustering effect that avoids the selection of a pre-defined number of clusters.

In this paper we propose an approach within the Bayesian paradigm to analyse the impact of multiple particle metrics on daily mortality, using the DP mixture model. Specifically, we provide a model that addresses, in a one-step procedure, both dimensionality reduction and regression. Our approach builds on the work of Molitor et al. (2010, 2011) which represents an alternative inferential approach to regression models when the covariates in analysis are correlated. The model, known as profile regression, performs a Bayesian clustering of the

covariates by identifying exposure profiles and, simultaneously, links these to a response variable in non-parametric form (even though the model continues to be parametric within clusters). Profile regression has found further applications in epidemiology and in genomics (Papathomas et al., 2011, 2012; Hastie et al., 2013). In this paper we extend this technique to analyse time series data, accounting for their typical features like trends, seasonality and temporal components through smooth functions. The resulting probabilistic solution groups time points with similar multipollutant and response profiles.

To demonstrate our approach, we used daily particle metric data from London (UK) 2002–2005 and daily number of deaths from respiratory diseases (Atkinson et al., 2010). Additionally, to assess the recent efforts in reducing air pollution in London, we also predicted a mean response profile for mortality in the year 2012. Using measurements collected at the same monitoring site, we compared the predictive distribution of mortality in 2012 against the one computed in 2005.

2. Material and methods

2.1. Description of the data

Atkinson et al. (2010) described results from an epidemiological time series study examining the effect of different metrics of particulate collected in London, on cardiorespiratory hospital admission and mortality using univariate log-linear Poisson models. We selected a subset of exposure data for the period January 2002 to December 2005 (years 2000–2001 were excluded due to poor data availability; for anions the proportion of missing data was about 96%), and respiratory-related mortality as the outcome. To predict respiratory mortality given the multipollutant scenario that London experienced in 2012, we measured the same set of particle metrics that were recorded in 2002–2005.

2.1.1. Mortality data

Daily count of deaths from respiratory diseases of London residents (2002–2005) were obtained from the Office for National Statistics and coded using the International Classification of Diseases, 10th Revision (ICD-10: Chapter J).

2.1.2. PM measurements 2002–2005

Daily average concentrations of particle metrics included: particle number concentration (PNC), inorganic anions such as chloride, nitrate and sulphate, black smoke (BS) and gravimetric measurements of PM, such as PM₁₀, PM_{2.5} and PM coarse fraction (that is, PM_{10–2.5} obtained by subtraction). With the exception of BS, the daily concentrations were obtained from a single background monitoring station in central London (North Kensington). BS was an average across several urban and suburban stations. PNC was measured using a TSI 3022A condensation particle counter, where particles are enlarged by condensation of saturated butanol vapour which are then counted using a laser and optical detector. The PM₁₀ 24-hour filter samples were collected at 16.7 l per minute on quartz fibre filters using Partisol 2025 (Thermo) instruments and these filters were analysed by ion chromatography. Finally, daily average gravimetric PM₁₀ and PM_{2.5} were sampled using a Partisol sampler and measured using methods in EN12341 and EN14907.

The data set also included PM apportioned into primary and non-primary sources (Fuller et al., 2002; Fuller and Green, 2006), giving modelled primary PM₁₀ (PPM₁₀), and non-primary PM subdivided by size fraction: non-primary PM₁₀ (NPPM₁₀), non-primary PM_{2.5} (NPPM_{2.5}), and non-primary PM coarse fraction (NPcoarse). The source apportionment model assumed that primary PM₁₀ was associated with nitrogen oxide (NO_x) sources and the non-primary component was the fraction of PM not associated with NO_x. NO_x is generally considered a robust marker for traffic pollution (Krzyzanowski et al., 2005).

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