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The effects of air mass transport, seasonality, and meteorology on pollutant levels at the Iskrba regional background station (1996–2014)

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HIGHLIGHTS

- Joint model of meteorology, seasons, trends & air mass movement gives more insight.
- Air mass from eastern Europe and northern Balkans leads to higher pollution levels.
- Decrease in trajectory length suggest a decrease in wind speeds in past 15 years.
- Bayesian approach to modelling leads to substantially easier-to-interpret results.

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ABSTRACT

Our main goal was to estimate the effects of long-range air transport on pollutant concentrations measured at the Iskrba regional background station (Slovenia). We cluster back-trajectories into categories and simultaneously model the effects of meteorology, seasonality, trends, and air mass trajectory clusters using a Bayesian statistical approach. This simplifies the interpretation of results and allows us to better identify the effects of individual variables, which is important, because pollutant concentrations, meteorology, and trajectories are seasonal and correlated. Similar to related work from other European sites, we find that slow and faster moving trajectories from eastern Europe and the northern part of the Balkan peninsula are associated with higher pollutant levels, while fast-moving trajectories from the Atlantic are associated with lower pollutant concentration. Overall, pollutant concentrations have decreased in the studied period.

1. Introduction

The presence of particulate matter (PM) and other pollutants in the urban environment is a health risk and long-term exposure can lead to respiratory and cardiovascular disease and other health-related issues (Bell et al., 2005; Feng and Yang, 2012; Hoek et al., 2013; Pope III and Dockery, 2006). Pollutant concentrations may vary on the hourly, daily, or weekly scales and are affected by seasonality and meteorology. It is also well established that aerosol components, such as those affecting PM concentrations, can be released at one location and travel to affect air quality at a different, possibly very distant location.

We focus on the Iskrba rural background station, a regional station for monitoring background air pollution. The main purpose of the study is to identify long-range transport patterns and measure their effect on local pollutant levels in Slovenia, while at the same time accounting for the effects of other relevant variables such as meteorology and seasonality.

Related studies of the effects of long-range transport on pollutant concentrations are extremely varied in terms of urban and rural locations and time periods. In terms of analytical methodology, however, these works can be split into two approaches. The first, and more commonly used approach, is to cluster air mass

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trajectories into distinct groups and then proceed with statistical analyses of the effects of these groups on pollutant concentrations (for some recent examples see, for example Dimitriou and Kassomenos, 2015; Donnelly et al., 2015; Makra et al., 2013; Paschalidou et al., 2015; Piñero-García et al., 2015). The second approach is attributing the changes in pollutant concentrations directly to grid-based locations based on trajectories that passed through those locations (for some recent examples see, for example Dimitriou and Kassomenos, 2015; Riuttanen et al., 2013; Tositti et al., 2013). Source attribution applications mostly follow or slightly modify the potential source contribution function of Ashbaugh et al. (1985) or the concentration field method by Seibert et al. (1994). For a broader review of methods for measuring the impact of air mass transport history on pollutant concentrations see Fleming et al. (2012).

In this study, we adopt the clustering-based approach. However, we deviate from the statistical analyses that typically follow once a set of clusters is determined. Related work relies on visual data exploration and null-hypothesis testing, either parametric (ANOVA) or non-parametric (Kruskal-Wallis), to inspect and test whether trajectories representing a cluster have an effect on local pollution levels or not. These approaches are limited: (a) they are adequate for measuring the presence of an effect, but not estimating effect size and (b) they make it difficult to quantify the uncertainty associated with the results. To deal with these shortcomings, we take a statistical (Bayesian) modelling approach. We jointly model and estimate the effects of air mass transport, meteorology, and seasonality. This novel methodology, in particular the unified approach to modelling effects on concentrations and the use of Bayesian statistics, and the substantive results that follow are the main contributions of this study.

The remainder of the paper is organized as follows. In Section 2 we describe the data, preprocessing steps, and our statistical methodology. In Section 3 we present and discuss the results of the analysis. With Section 4 conclude the paper and provide some directions for future work.

2. Materials and methods

In this section we first describe the data and preprocessing then proceed with defining the statistical methodology.

2.1. Data and preprocessing

The Iskrba monitoring station was established as part of the EMEP (European Monitoring and Evaluation Programme). Designated as a rural background site under EU Directive 2008/50/EC, the stations location was chosen so that the effects of local pollution sources are negligible, which makes it appropriate for studying long-range transport of pollutants. It is located at 45.57°N, 14.87°E on a plateau at approximately 500 m a.s.l. in a prevalently woody area and within a transitional climate zone.

Most pollutant level measurements at the Iskrba station are based on standard EMEP 24-h periods, starting at 08:00 CET. We preprocessed all the data to fit within that 24-h frame, starting at 08:00 CET and ending at 08:00 CET the next day. That is, each sample from the processed data represents one 24-h period. The details on how the pollutant, meteorological, seasonality, and air flow data were preprocessed and included are found below.

2.1.1. Pollutant level data

We included several different pollutants as measured at the Iskrba station by various methods until December 31, 2014 (see Table 1 for a summary). Measurements were made following the guidelines and standards set in the EMEP Manual for Sampling and Analysis.[1] Measuring instruments were regularly calibrated. Certified reference materials were used in sample analyses and the laboratory regularly participates in inter-laboratory comparison schemes.

Table 1
Summary of pollutant concentrations. Note that pollutant concentration levels and seasonality patterns at the Iskrba have been consistent throughout the observed period, with the exception of a slight downwards trend in some pollutants as reported in the results.

[in $\mu g/m^3$]	Median	5th percentile	95th percentile	N
O_3^{max}	90.5	41.35	140.67	4593
PM_{10}	13.24	4.78	29.83	3489
$PM_{2.5}$	9.94	3.38	25.21	3127
NO_2	0.43	0.21	1.27	3413
SO2	0.27	0.03	2.57	6538
SO_4^{2-}	0.65	0.1	2.33	6604
$HNO_3+NO_3^-$	0.18	0.04	0.74	6596
$NH_3+NH_4^+$	0.84	0.19	2.35	6600

SO_2, SO_4^{2-}, $HNO_3+NO_3^-$, and $NH_3+NH_4^+$ were collected by filter-pack sampling system (NILU EK sequential sampler) at external conditions for 24-h periods starting at 08:00 CET. A three-stage filter pack system was used (Teflon filter, filter impregnated with KOH, and filter impregnated with oxalic acid). Samples were analysed in a laboratory setting using ion chromatography (Shimadzu CDD-10Avp and Dionex DX-120). These data were available from May 15, 1996.

NO_2 has been sampled at room temperature and humidity using sodium-iodide impregnated sintered glass filters since April 14, 2004 for 24-h periods also starting at 08:00 CET (NILU SS2000 sequential sampler) and analysed in a laboratory setting with a spectrometer (Agilent cary 50).

PM_{10} and $PM_{2.5}$ data were sampled with the same 24-h periods starting at 08:00 CET and are performed every day since 2005 for PM_{10} and since 2006 for $PM_{2.5}$. Samples were obtained at external conditions with sequential sampling (Sven Leckel SEQ47/50) with automatic filter changer and analysed using the standard gravimetric method after being conditioned for 48 h at 20 °C ± 1 °C, and 50% ± 5% relative humidity.

Ozone measurements for the station Iskrba were performed since 2002 on hourly basis using ultraviolet photometry (Thermo Scientific 49c ozone analyser). In line with ozone concentration reporting in EU legislature, we also focus on the maximum recorded value in the 24-h period.

Detection limits were 0.003 $\mu gN/m^3$ ($HNO_3+NO_3^-$), 0.001 $\mu gS/m^3$ (SO^2), 0.006 $\mu gN/m^3$ ($NH_3+NH_4^+$), 0.003 $\mu gS/m^3$ (SO_4^{2-}), 0.29 $\mu g/m^3$ (PM^{10}), and 0.35 $\mu g/m^3$ ($PM^{2.5}$). Combined measurement uncertainty (coverage factor $k = 2$) for determination of PM10 by gravimetric method SIST EN 12341:2014 is 12.5%.

2.1.2. Meteorological data

Meteorological data were collected by automatic weather station (AWS) in 30-min intervals. In our analysis we use mean temperature and mean humidity at 2 m above ground, mean wind speed at 10 m above ground, and precipitation amount. We aggregate the data based on the time frame between 08:00 CET and 08:00 CET the next day to match 24-h pollutant concentration measurements periods. That is, we split the data into subsets according to 24-h pollutant measurements period instead of starting at midnight. Some of the meteorological measurements have skewed distributions. After aggregation, we log-transform wind speed and logit-transform ($log\left(\frac{x}{1-x}\right)$) humidity to obtain

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[1] http://www.nilu.no/projects/ccc/manual/index.html