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METEOROLOGICALLY ADJUSTED GROUND-LEVEL OZONE TREND AT THE PREILA BACKGROUND SITE

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Using a multiple low-pass Kolmogorov–Zurbenko filter, the separation of the original signal based on ozone and temperature data into long-term (annual), seasonal, and short-term (synoptic scale) components was applied for removing the effects of meteorological factors at the Preila background station (Lithuania). Meteorologically adjusted hourly ozone concentration data revealed long-term variations. Based on this approach a decline of 0.36 μg m^{-3} yr^{-1} in the meteorologically adjusted ozone concentration was determined for the period of 1999–2004.

Keywords: ozone concentration, time-series, mathematical statistics, Kolmogorov–Zurbenko low-pass filter

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1. Introduction

Surface ozone levels and their changes are of great interest since positive trends of its concentration have been established at various sites [1, 2]. The ozone level variations and its trend mainly depend on meteorological conditions, precursor emission characteristics, and the concentration of different pollutants.

The recent decade has seen a growing variety of statistical analysis on the subject applying a broad range of statistical methodologies with adjustment being considered for different policy objectives. The meteorological adjustment of ozone trend can be achieved by statistical modelling of the relationship between ozone concentration and meteorological variables.

The air quality is influenced by a variety of processes, including anthropogenic and biogenic emissions. However, changes caused by emissions are often difficult to detect in the air quality parameter record due to the prevailing influence of climate and weather conditions, which have the greatest impact on daily variations. Unless the change in emissions is substantial, the resulting improvement in the ozone air quality can be easily masked by the meteorological variability. The presence of various scales of motion in time-series can complicate the analysis and interpretation of long-term trends in meteorological variables. Thus, the influence of weather variability must be removed in order to assess changes due to emissions [3].

Time-series of atmospheric pollutant concentrations contain fluctuations occurring on many different time scales. Spectral analysis indicates that the largest forcing in the hourly ozone time-series data is the diurnal one having a period of 24 hours [4]. Because of its large influence, it is necessary to separate the diurnal signal from the time-series. Additional frequency bands of interest are the intra-day range, the synoptic range, and longer-term fluctuations (i. e., baseline).

In this study, the Kolmogorov–Zurbenko low-pass filter (KZ filter) was used to separate ozone and temperature data into short-term, seasonal, and long-term trend components in order to identify long-term trends in the ozone concentration.

2. Methods

The application of statistical methods to the analysis of temporal variations is demonstrated using hourly ozone concentrations recorded at the coastal background site. The Preila environmental pollution research station (55°20′ N, 21°00′ E, 5 m a.s.l.) is located in western Lithuania on the coast of the Baltic Sea, on the Curonian Spit. Since the Preila station is located far from substantial local sources of airborne pollutants, it

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can be characterized as a regionally representative background site for Lithuania.

The measurement data cover the period from 1999 to 2004. Time-series of ozone and temperature measurement data contain fluctuations occurring on different time scales [5, 6]. Since ozone measurements are performed at discrete intervals, the highest and lowest frequencies that can be estimated for any particular time-series are determined by the sampling interval and the length of data record, respectively. The choice of different frequency bands used by Hogrefe et al. [7, 8] and Biswaset et al. [9] was based on the recorded power spectrum and on *a priori* knowledge about different physical processes.

Changing meteorological conditions due to the presence of nearly stagnant high-pressure system or the passage of frontal systems cause the variations of ozone on the synoptic scale. Fluctuations of the baseline are expected to be caused by such processes as seasonal variations of the solar flux changing large-scale flow patterns, and the change in vegetation coverage and biogenic emissions. The separation of the natural logarithm of ozone time-series into distinct components is achieved by the application of the Kolmogorov–Zurbenko filter [8]. The KZ filter isolates component frequencies by repeated iterations of a simple, point-centred moving average. Hogrefe et al. [7, 8] and Biswaset et al. [9] used the Kolmogorov–Zurbenko filter because of its powerful separation characteristics, simplicity, and ability to handle missing data.

According to [10], the components of the ozone time-series to be separated and analysed are defined by

$$O_3(t) = E(t) + S(t) + W(t)\,, \tag{1}$$

where $O_3(t)$ is the natural logarithm of the original ozone time-series, $E(t)$ is the long-term or trend component, $S(t)$ is the seasonal change, and $W(t)$ is the short-term variation.

The deterministic portions (E and S) produced by repeated iterations of a simple moving average are separated from the short-term variations in the data using the KZ filter. Each iteration of the moving average is defined by

$$X_t^{i+1} = \frac{1}{m} \sum_{j=-k}^{k} X_{t+j}^{i}\,, \tag{2}$$

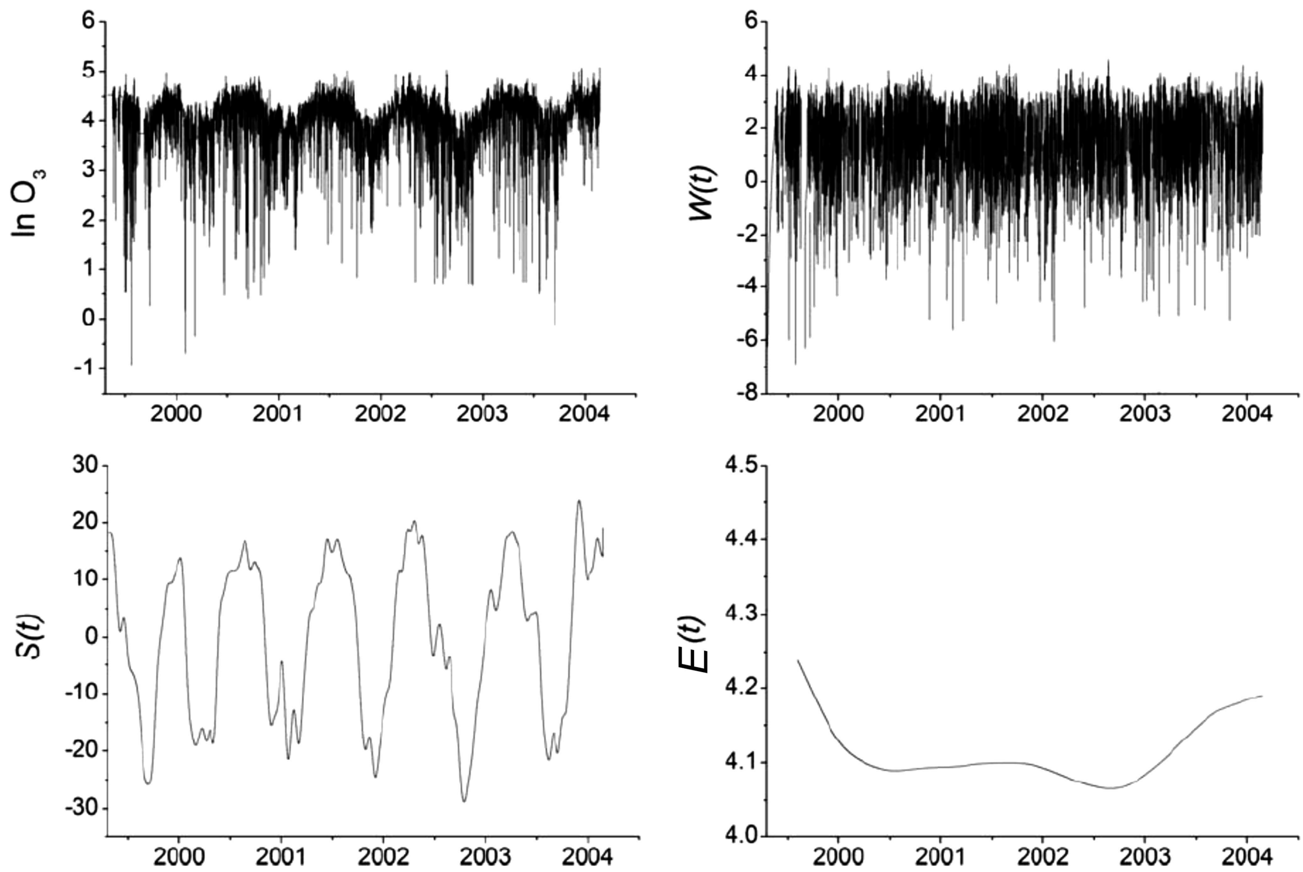

Fig. 1. Filtered ozone data: $O_3(t)$ (upper left), $W(t)$ (upper right), $S(t)$ (not log-transformed) (lower left), and $E(t)$ (lower right) components.

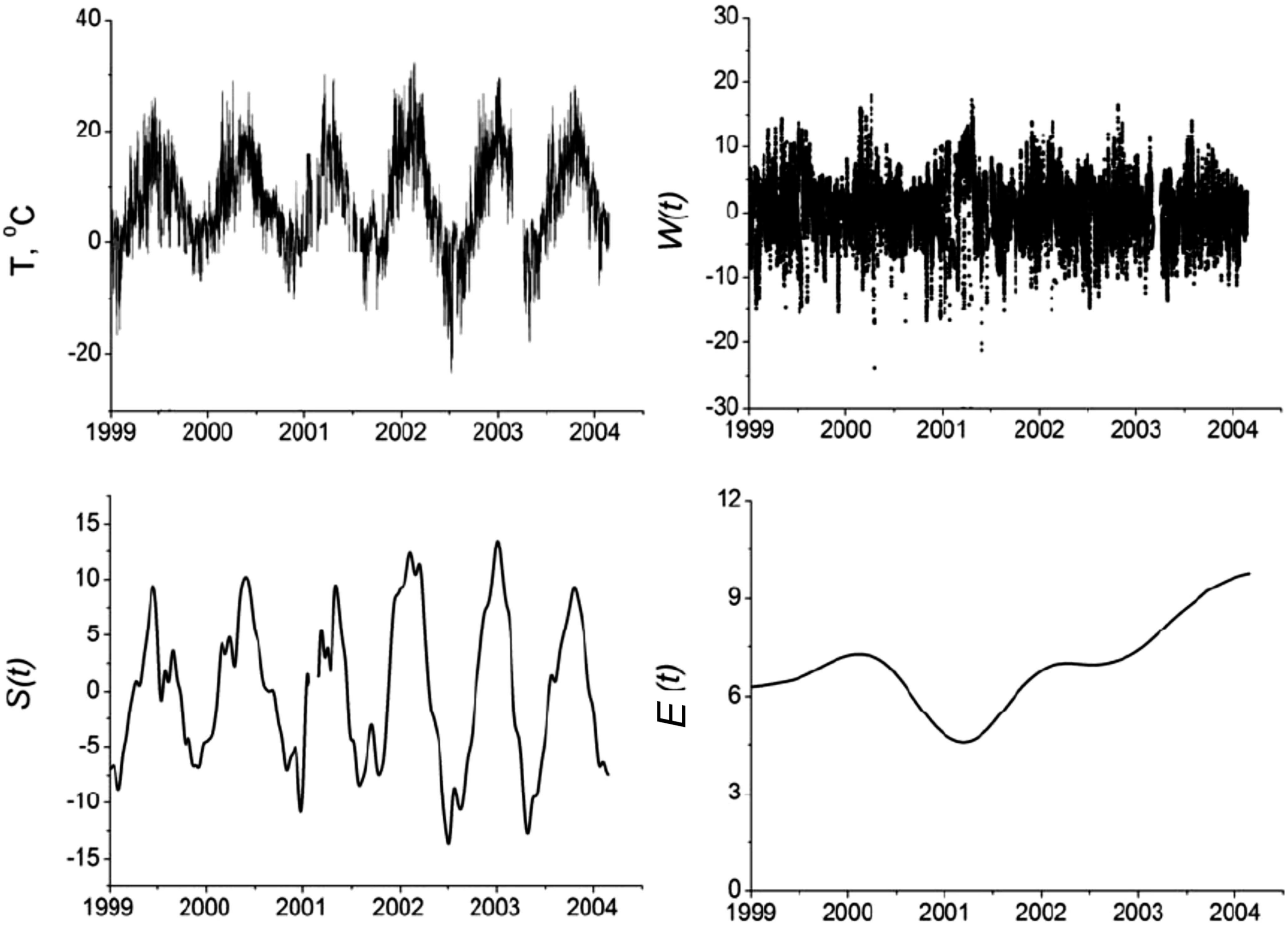

Fig. 2. Filtered temperature data: T, °C (upper left), $W(t)$ (upper right), $S(t)$ (lower left), and $E(t)$ (lower right) components.

where $X_t^0 = X_t$, $m = 2k + 1$.

It can be written that

$$KZ_{m,p}(X_t) = X_t^p\,, \tag{3}$$

where $KZ_{m,p}$ is the KZ filter with a window size of m days and p iterations.

The temporal components are estimated as follows:

$$S(t) = KZ_{15,5}(O_3(t)) - KZ_{365,3}(O_3(t))\,, \tag{4}$$

$$E(t) = KZ_{365,3}(O_3(t))\,. \tag{5}$$

The $KZ_{15,5}$ low-pass filter was applied to the log-transformed ozone and temperature values yielding the baseline time-series presented in Figs. 1 and 2. It is clear from these plots that short-term cycles have been removed. The residuals are the seasonal cycles and the long-term variations.

The resulting data set (which is a data set of residuals or "differences" contains the trend due to changes in control strategies, emissions of ozone precursors, as well as any year-to-year changes in weather and climate not captured in the temperature time-series.

3. Results

Using two KZ filters – a 15-day average, 5-pass filter and a 365-day average, 3-pass filter, the time-series of ozone concentrations were separated into a long-term component representing the influence of trends in precursor emissions, a seasonal component representing the influence of the Earth's rotation about the Sun, and a short-term component representing the influence of fluctuating synoptical-meteorological conditions and random processes (noise). An analysis of variance has shown that a relative contribution of the components of the ozone time-series to the total variation of the data set is different. Long-term component frequency variability was 2.3%, the seasonal component frequency variability contribution was 19.0%, while the median short-term component was 26.4%. The results illustrate that the intra-day component is the largest contributor to the overall variance in observations (52.3%).

An examination of the 1999–2004 record of ozone data indicated that the annual mean ozone concentration was decreasing by 1.2 μg m^{-3} per year. However, the meteorological variability could mask the actual

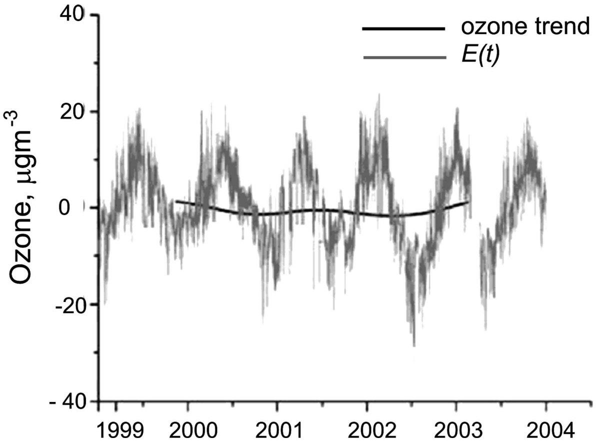

Fig. 3. Resulting total residuals after regression of ozone baseline and short-term component, and long-term component.

trend. Thus, the Kolmogorov–Zurbenko meteorological trend decomposition approach was applied separately to ozone data to comply with the meteorologically independent ozone time-series. Data sets are plotted in Figs. 1 and 2, respectively.

Generally, ozone concentrations well correlate with temperature. Because of this relationship, temperature data can be used to remove the weather-related variability in the ozone time-series.

The meteorologically adjusted ozone trend was obtained through regressions between the components of the ozone and temperature baseline and short-term components. The resulting total residuals were removed from the ozone long-term component ($KZ_{365,3}$ data set) (Fig. 3).

The trend identified by the slope of a regression line reflects long-term changes in emissions and can be used to assess progress and to predict a future improvement in the air quality under the assumption that the trend will continue. The slope is significant at the 95% confidence level and yields a projected reduction of 0.36 μg m^{-3} per year, 1999–2004. A smooth curve in Fig. 3 shows the effects of long-term changes in emissions and the aspects of climate and weather that are not well-represented by temperature alone.

The long-term trend was analysed to determine if there was a significant ozone concentration decrease over the period of the study. Smoothed residuals represent changes in the ozone concentration attributable to sources other than the removed parameter of the temperature component, such as emissions or long-term climate changes.

4. Conclusions

In this work, using the Kolmogorov–Zurbenko low-pass filter, the original time-series consisting of the logarithm of hourly ozone concentrations and temperature measured at the Preila background site from 1999 to 2004 were split into long-term, seasonal, and short-term components. The analysis demonstrated that meteorological effects influenced the trends in the ozone concentration. Time scale separations were based on diurnal, synoptic, seasonal, and long-term fluctuations. The filtered time scales are compared to show the contribution of individual time scales to the total variability of ozone at the site. The intra-day and short-term, high frequency variability has a higher percentage contribution to the overall ozone variability at the site (52.3% and 26.4%, respectively).

These results showed a strong meteorological influence on the ozone time-series. The overall ozone trend showed a negative change of 1.2 μg m^{-3} yr^{-1}, although the meteorologically adjusted trend significant at the 95% confidence level showed a decrease of 0.36 μg m^{-3} yr^{-1} for the period of 1999–2004. This trend could result from changes in overall emissions, pollutant transport, climate, policy, and economic factors.

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PRIEŽEMIO OZONO KONCENTRACIJOS PREILOS STOTYJE KITIMO TENDENCIJOS KOREKCIJA DĖL METEOROLOGINIŲ VEIKSNIŲ ĮTAKOS

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Santrauka

Pateikiama ozono koncentracijos Preilos foninėje stotyje 1999–2004 metais kitimo priklausomybės nuo meteorologinių veiksnių analizė. Pritaikius Kolmogorovo ir Zurbenkos žemų dažnių filtrą, ozono koncentracijos ir temperatūros laiko eilutės originalus signalas buvo suskaidytas į skirtingus sandus. Nustatyta ozono koncentracijos mažėjimo (0,36 μg m^{-3} per metus) tendencija, neatsižvelgiant į meteorologinių veiksnių įtaką. Taip pat nustatyti laiko eilutės dažnių kintamumo dėl meteorologinių veiksnių indėliai, lemiantys ozono lygio svyravimus. Didžiausią įtaką ozono lygio svyravimui turėjo meteorologinių veiksnių paros kitimas (52,3%), sezoninis (19%), trumpalaikis (26,4%) ir ilgalaikis (2,3%) kitimai.