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Trends of primary and secondary pollutant concentrations in Finland in 1994–2007

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ABSTRACT

The trends in the atmospheric concentrations of the main gaseous and particulate pollutants in urban, industrial and rural environments across Finland were estimated for the period of 1994–2007. The statistical analysis was based on generalized least-squares regression with classical decomposition and autoregressive moving average (ARMA) errors, which was applied to monthly-averaged data. In addition, three alternative methods were tested. Altogether 102 pollutant time series from 42 sites were analyzed. During the study period, the concentrations of SO_2, CO and NO_x declined considerably and widely across Finland. The SO_2 concentrations at urban and industrial sites were approaching background levels. The reductions in NO_x and CO concentrations were comparable to those in national road traffic emissions. A downward trend was detected in half of the NO_2 time series studied, but the reductions were not as large as would be expected on the basis of emission trends, or from NO_x concentrations. For O_3, neither the mean nor peak values showed large changes in background areas, but were increasing in the urban data. For PM_{10}, five of the 12 urban time series showed decreasing mean levels. However, the highest concentrations, typically attributable to the problematic springtime street dust, did not decrease as widely. The reduction of the long-range transported major ions, mainly driven by the large-scale reduction in sulphur emissions, possibly plays a significant part in the decreases in the mean PM_{10} concentrations. It was shown that the handling of the serially-correlated data with the ARMA processes improved the analysis of monthly values. The use of monthly rather than annually-averaged data helped to identify the weakest trends.

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1. Introduction

Within European-wide assessments of air quality, Finland and other Nordic countries are repeatedly highlighted as those least affected by local and regional air pollution (e.g. EEA, 2003, 2007). For Finland, there are several obvious reasons behind this position. Firstly, its location in northernmost Europe, separated from the main continent by the Baltic Sea, reduces the transboundary influence of the major European source areas. Secondly, the population of about 5.3 million people spread over 338,000 km^2 limits the size of cities, and makes Finland one of the most sparsely-populated countries in Europe. Thirdly, as a response to the threat of forest and lake acidification in the late 1970s and the 1980s, considerable emission abatement measures were accomplished within energy production and industry in the 1980s, which also improved local-scale air quality. Moreover, the previous decades had already witnessed a widespread introduction of district heating from combined heat and power plants, which together with increasing stack heights evidently had a positive effect on urban air quality.

The concentrations of most air pollutants are low in Finland as compared to many regions in Europe. For example, the occurrences of values over the limit, threshold and target concentrations set by the European Union (EU) are very rare. However, this does not mean that air pollution no longer poses any potential threats. For example, the background concentration of ozone is high, even in the remote northern parts of the country (e.g. Laurila et al., 2004, 2009); in urban areas, including minor towns, local concentrations of particulate matter (PM) may persist at highly elevated levels for weeks during the so-called springtime dust period (Anttila and Salmi, 2006); small-scale wood combustion increases wintertime concentrations of benzo(a)pyrene in residential areas (Hellén et al., 2008); episodic transboundary pollution from wild fires may cause high PM concentrations over large areas, potentially enriched with carcinogenic polycyclic aromatic hydrocarbons (e.g. Anttila et al., 2008; Niemi et al., 2009). Consequently, while the 1980s can be characterized as a decade of successful reduction of sulphur pollution from energy production and industry, in the 1990s and

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2000s it has been necessary to focus attention on ozone, its nitrogen precursors (NO, NO_2) and PM, on account of their adverse effects on human health and ecosystems. Since joining the EU in 1995, Finland has followed the emission control framework developed within the EU. Now, more than ten years later, it seems appropriate to evaluate the development of air quality in Finland during this period.

The objective of this study is to quantify trends in the atmospheric concentrations of the main gaseous and particulate pollutants from 1994 to 2007, to briefly compare these with national emission trends and to discuss the effectiveness of emission control measures. Air pollution trends are estimated for NO_2, NO_x ($NO + NO_2$), O_3, SO_2, CO, PM_{10} (particles of less than 10 μm in aerodynamic diameter), SO_4^{2-} (p), NO_3^- (p) + HNO_3 (g) and NH_4^+ (p) + NH_3 (g) concentrations, measured in various locations across Finland. This is the first national trend study that extensively covers both urban and industrial environments as well as the rural background. The statistical analysis is based on an advanced method that evaluates the serial correlation in a time series, but we also test three other statistical techniques for detecting linear trends.

2. Data

The measurement data used in this study were downloaded from the national air quality database maintained by the Finnish Meteorological Institute (FMI). Air quality monitoring data from all Finnish urban, industrial and background networks are collected annually into this database, which contains a total of 30 networks with about 120 permanent measurement sites. The more comprehensive hourly data, measured with common commercial automatic analyzers, are available from this database from 1994, which was taken as the start year of this study.

The networks themselves are responsible for the quality assurance of their measurements. Nationwide field audits of the operations and quality control systems of the networks, completed with field comparison campaigns, were performed by FMI in 2002–2003 and 2006 (Waldén et al., 2004, 2008). In both comparisons, the results of all the networks were very good for O_3, SO_2 and CO, the deviation from the reference data in nearly all cases being less than 8%, which was set as the limit value for an acceptable result. For NO, which is the compound detected by NO_x analyzers, the results of the former comparison were considered satisfactory, with two-thirds of the results lying within the 8% deviation (Waldén et al., 2004), while in the latter there was only one anomalous observation (Waldén et al., 2008).

The PM measurements have not yet been nationally intercompared. The mass concentrations of PM_{10} presented in this study are measured either by the tapered element oscillating microbalance or the beta-attenuation method. Neither of these methods is necessarily fully compatible with the gravimetric methods, nor with each other, due to the unknown losses of semi-volatile compounds. In this study, we did not apply any corrections for these losses, but included in the trend analysis only those sites at which the PM measurement method had remained the same during the whole study period.

Within the database, the stations are qualitatively classified as urban, suburban or rural according to the type of site surroundings, and as traffic, industry or background in relation to the dominant emission source, in accordance with the European Commission decision 2001/752/EC (EC, 2001). A summary of the measurement sites and components included from each site is presented in Table 1.

The monthly and annual means of NO_2, NO_x, O_3, SO_2, CO and PM_{10} were calculated from the hourly data for the years 1994–2007 for all available monitoring stations. A data completeness of 75% was required for an acceptable monthly value. For further processing, only those time series with at least ten years of valid data between 1994 and 2007 were accepted. Single missing monthly values in the middle of time series (up to a maximum of five per year and site) were replaced by the mean of all accepted values for the month and site in question. In two cases (the Jyväskylä Lyseo and Oulu Keskusta stations), the time series were formed from the combination of two subsets of data, resulting from a minor transfer (a couple of hundred metres) of the site location. In these cases, the hourly and daily data were also visually inspected to ensure that there was no stepwise change in the measured concentrations.

In addition to the continuously-analyzed components listed above, analogous monthly time series were compiled from the daily atmospheric concentrations of sulphate (SO_4^{2-} (p)), nitrate (representing the sum of NO_3^- (p) and HNO_3 (g)) and ammonium (the sum of NH_4^+ (p) and NH_3 (g)) measured at three EMEP (European Monitoring and Evaluation Programme) stations: Korppoo Utö, Kuusamo Oulanka and Virolahti. For these sites, SO_2 concentrations measured with the filter pack technique (Leppänen et al., 2005) were also utilized. The data from the EMEP stations were available up to 2006. Altogether 102 pollutant time series from 42 sites passed the validity criteria (Table 1).

3. Statistical methods

Four different methods were used for estimating the linear trend in the concentration time series: (1) Generalized Least-Squares (GLS) regression with classical decomposition and autoregressive moving average (ARMA) errors, applied to monthly data; (2) Ordinary Least-Squares (OLS) regression applied to deseasonalized monthly data; (3) OLS regression applied to annual data; (4) Non-parametric Sen's slope estimation method with the non-parametric Mann–Kendall significance test, applied to annual data. The GLS-ARMA-based method was used as the reference method, with which the other methods were compared. In this statistical model, the autocorrelation typically present in air pollution concentration time series is accounted for by iteratively applying an ARMA-based correlation structure to the residuals of the fitted linear model.

The seasonal decomposition employed for the methods (1) and (2) above was carried out following Brockwell and Davis (2002). Briefly, the monthly time series were deseasonalized by the classical seasonal decomposition, in which a moving average with a 13-month window (with the first and last observations averaged) was first subtracted from the original data. Next, the mean of the monthly deviations thus obtained was subtracted, to ensure that the mean of the seasonal component is zero. The seasonal component was then calculated by averaging the scaled deviations on a monthly basis. Finally, a deseasonalized time series was produced by subtracting this seasonal component from the original data.

The GLS-ARMA-based trend analysis consisted of the following steps; for additional details, see Brockwell and Davis (2002).

(*i*) A first-order OLS regression model was fitted to the deseasonalized data.

(*ii*) The autocorrelation function (ACF) of the resulting residuals (i.e., the noise obtained by subtracting the estimated seasonal and trend components from the original time series) was calculated. If 5% of the values of the sample ACF up to a lag of 40 were outside the bounds $\pm 1.96/\sqrt{n}$ (corresponding to the 95% confidence interval), where n is the number of observations, it was concluded that the residuals were not independent and identically-distributed (i.i.d.) random variables, and an ARMA model was introduced.

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