## MODELLING AIR POLLUTION DATA IN COUNTRYSIDE AND URBAN ENVIRONMENT, HUNGARY

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## ABSTRACT

The aim of the study is to determine spatial and temporal characteristics, as well as statistical interrelationships of air pollutants in Szeged and Csongrád county and to give human biometeorological assessment on air pollution load there. Monthly averages of NO<sub>2</sub>, SO<sub>2</sub> and deposited dust from RIE (Regional Immission Examining) network operating in Szeged (10 stations) and Csongrád county (11 stations), furthermore 30-minute averages of CO, NO, NO<sub>2</sub>, SO<sub>2</sub>, O<sub>3</sub> and total suspended particulate (TSP) measured by the monitoring station in Szeged are analysed. Emission time courses of the pollutants and temporal variability of TSP are shown. Joint daily behaviour of O<sub>3</sub>-irradiation, O<sub>3</sub>-NO and NO<sub>2</sub>-irradiation are presented. Spatial factor analysis is used for NO<sub>2</sub>, SO<sub>2</sub> and deposited dust time series to produce their homogenous subregions. Air quality stress index for mean (annual) and short-term (diurnal) air pollution load was calculated. Average diurnal cycle of the pollutants in percentage of the health limits are also displayed.

**Key words:** emission time courses, temporal variability, joint daily behaviour, spatial factor analysis, air pollution load

## **1. INTRODUCTION**

Air pollution is one of the most important environmental problems, which concentrates mostly in cities. Generally, human activities induce monotonous accumulation of pollutants. Possible reasons of worsening air quality are population growth in cities and, in connection with this, increasing built-in areas there. A considerable part of population growth is coming from the migration to the cities. The ever-increasing urban population, together with the growing industrialisation and energy consumption and the extensive transportation, raise air pollution, which becomes a more and more serious challenge for the interest of survival.

The aim of the study is to determine spatial and temporal characteristics, as well as statistical interrelationships of air pollutants in Szeged and Csongrád county (Fig. 1.) and to give human biometeorological assessment on air pollution load there.



Geographical position of Fig. 1. the monitoring and RIE stations in Csongrád county and Szeged (top left) with built-in types of the city (a: centre (2-4-storey buildings); b: housing estates (5-10-storey buildings); c: detached houses (1-2-storey buildings); d: industrial areas; e: green areas; (1): monitoring station, (2): measurements for  $NO_2$ ,  $SO_2$  and deposited dust, (3): measurements for only deposited dust, (4): measurements for only NO<sub>2</sub> and SO<sub>2</sub>.

The database of the study results, on the one hand, from the monitoring station in Szeged downtown, for the period between 1997-2000. This station is located at a busy traffic junction and measures mass concentrations of CO, NO, NO<sub>2</sub>, SO<sub>2</sub>, O<sub>3</sub> and TSP (Fig. 1.). Another source of the database is the RIE (Regional Immission Examination) network for Szeged and Csongrád county. From this network, we used monthly mean concentrations of NO<sub>2</sub> and SO<sub>2</sub>, as well as monthly totals of deposited dust (g /  $m^2$  / number of days of the month) for the period between 1985-1999. Szeged is located in the Carpathian Basin, at 20°06'E and 46°15'N, at an altitude of ca. 80 m above sea level. Its population is about 155,000 inhabitants and the built-in area is around 46  $\text{km}^2$ . The city is situated near the confluence of the Tisza and Maros Rivers, in southern Hungary. Szeged is the largest city and the centre of light industry in the southern part of the Great Hungarian Plain (Fig.1.).

The industrial area is located in the north-western part of Szeged. Thus, the prevailing westerlies and northerlies transport pollutants, from this area, towards the centre of the city.

### 2. MAIN TEXT

2.1. Statistical methods 2.1.1. Factor analysis

We investigated summer and winter half-year data sets of  $NO_2$ ,  $SO_2$  and deposited dust both for Szeged and the countryside stations with factor analysis of the spatial variability. Our aim was to identify subregions with more or less sovereign variations of these parameters.

One of the best methods to study time series data for a large number of stations or grid points, where strong spatial and temporal correlation prevails, is *factor analysis* (see e.g. Bartzokas and Metaxas, 1993). One of the main benefits of this method is the reduction of the initial variables into much fewer uncorrelated ones, namely the factors. In this way, regions can be defined where, for any point within each region, the analysed meteorological variable covaries. Each original variable,  $P_i$ , i = 1, 2, ..., n, can be expressed as  $P_i = a_{i1}F_1 + a_{i2}F_2 + ... + a_{im}F_m$  (m < n), where  $F_j$ , j = 1, 2, ..., m, are the factors and  $a_{ij}$  are the loadings. One important stage of this method is the decision for the number (m) of the retained factors. On this matter, many criteria have been proposed. In this study, the *Guttman criterion* or *Rule 1* is used, which determines to keep the factors with eigenvalues > 1 and neglect the ones that do not account for at least the variance of one standardised variable. Another vital stage in this analysis is the so-called rotation of the axes (factors). This process achieves discrimination among the loadings making the rotated axes easier to interpret. In this analysis the *Orthogonal Varimax Rotation* was applied, which keeps the factors uncorrelated.

#### 2.1.2. Numerical expression of air pollution load

The air quality stress index (AQSI) can be determined for mean (annual) and short-term (diurnal) air pollution loads. It considers only the following components: sulphur dioxide (SO<sub>2</sub>), nitrogen dioxide (NO<sub>2</sub>) and total suspended dust (TSP) (Mayer, 1995). The air quality stress index (further: AQSI) for mean (annual) air pollution (AQSI<sub>1</sub>) is given as follows:

$$\begin{array}{ll} AQSI_1 = 1/3 \cdot [I1(SO_2)/50 + I1(NO_2)/50 + I1(TSP)/50] & (1) \\ The AQSI for short-term (diurnal) air pollution (AQSI_2) is given below (after Mayer, 1995): \\ AQSI_2 = 1/3 \cdot [I2(SO_2)/125 + I2(NO_2)/150 + I2(TSP)/150] & (2) \end{array}$$

#### 2.2. Results

#### 2.2.1. Emission time courses of air pollutants

Emissions of air pollutants can be originated from different source groups. These groups are as follows: residential, services, transport, power plants, other heating, industry and agriculture. The environmental station in Szeged, placed at a busy traffic junction, monitors and stores concentrations of CO, NO, NO<sub>2</sub>, O<sub>3</sub>, SO<sub>2</sub> and TSP. Among them, CO, NO, NO<sub>2</sub> and O<sub>3</sub> are more important because of their role in traffic. Average diurnal course of them shows a definite picture with double wave for CO, NO and NO<sub>2</sub> and one wave for O<sub>3</sub>. They all can be connected to traffic density. Opposite daily course of NO and O<sub>3</sub> is also very clear.

The most important air pollutants are nitrogen oxides  $(NO_x)$ , sulphur dioxide  $(SO_2)$ , total suspended particulate (TSP) and carbon monoxide (CO). Their trends are presented in Fig. 2.



Fig. 2. Emissions of nitrogen oxides  $(NO_x)$ , sulphur dioxide  $(SO_2)$ , total suspended particulate (TSP) and carbon monoxide (CO) in Szeged and Csongrád county from 1989 until 1999 (after the Environmental Protection Inspectorate of Lower-Tisza Region, Szeged, 2001).

Only  $SO_2$  emissions show significant decrease both in Szeged and Csongrád county. This might be explained by the change of fuel pattern and technology after1990. Concentration of the other three air pollutants displays no temporal change. During the last decade (1991-2000), many advances have been achieved in the field of transportation, such as the widespread use of unleaded petrol, enforcement of stricter vehicle emission standards and the growing ratio of more recent vehicle models. Hence, the increasing traffic density might be the reason of stagnating or even increasing CO and  $NO_x$  concentrations in the region.

Transport, especially with  $NO_x$ - and CO-emissions, can be considered a substantial pollution source. In Hungary, emissions of CO and  $NO_x$  caused by motor vehicle traffic amount to 44% and 53% of the total, respectively. Consequently, motor vehicle traffic seems to be one of the most important sources of air pollution, mainly in cities. Szeged, together with the population and the number of motor vehicles, is continuously growing. Hence, perspectives indicate a more significant role for traffic.

#### 2.2.2. Temporal variability of total suspended particulate (TSP)

The average annual cycle of total suspended particulate shows the greatest values in November, December and January, with its maximum in January (Fig. 3.). Higher winter values might refer to atmospheric stability with frequent inversions. The lowest values in summer (June, July, August and September) can be explained by dilution because of intensive vertical exchange in the atmosphere. The diurnal and weekly cycle of total suspended particulate (TSP) (Fig. 4.) has the shape of a double wave. Both primary and secondary maxima can be observed during peak hours and, in the same way, primary and secondary minima occur, when traffic is least (at night) or decrease (around midday). Also, due to the traffic density, the concentration of TSP is relatively higher on weekdays, than on weekends.



Fig. 3. Average annual cycles of TSP at the monitoring station, Szeged downtown, for the period 1997-2000.



Fig 5. Average weekly and diurnal cycle of percentile values of TSP at the monitoring station, Szeged downtown, for the period between 1997-2000.



Fig. 4. Average weekly and diurnal cycles of TSP at the monitoring station, Szeged downtown, for the period 1997-2000.

Peak values for TSP show a double wave displaying maxima late in the evening, while secondary maxima can be observed late in the morning. Least TSP concentrations are measured early in the mourning, whilst secondary minima occur in the evening (Fig. 5.). The average annual, weekly and diurnal cycles, as well as average weekly and diurnal cycle of percentile values of TSP are very similar to those of NO, which highlights connection of TSP with traffic (Makra et al., 2001).

#### 2.2.3. Joint daily behaviour of pairs of variables

In order to receive more detailed information on interrelationships among  $O_3$ , NO, NO<sub>2</sub> concentrations and irradiation, joint daily behaviour of their pairs were studied (Fig. 6.). In the graphs, serial numbers of the measurements are presented. (No. 1: value of the measurement at  $0^{30}$ , ..., No. 48: value of the measurement at  $24^{00}$ .) O<sub>3</sub>-light pair shows a clear linear connection with some lag, while both O<sub>3</sub>-NO and NO<sub>2</sub>-light pairs basically display an opposite connection.





Joint daily behaviour of the pairs can be explained as follows. Ratio of NO<sub>2</sub>/NO

depends, through ozone concentration, on radiation (on the figure: light) and NO

emissions. Daytime, the ratio  $NO_2/NO > 1$ can be explained by the rapid oxidation of  $NO (NO + O_3 \longrightarrow NO_2 + O_2)$  (intensive, ozone producing processes). While the turn of this ratio in the evening and at night indicates decrease of oxidation capacity of the atmosphere (following nightfall, photochemical processes, leading to ozone formation, stop) (Horváth et al., 2001).

Fig. 6. Joint daily behaviour of pairs of variables ( $O_3$ , NO, NO<sub>2</sub>: ppb; light: W m<sup>-2</sup>)

## 2.2.4. Spatial classification

Kholmogorov-Smirnov test was applied for data sets of NO<sub>2</sub>, SO<sub>2</sub> and deposited dust in Szeged and Csongrád county, according to which all of them proved to be of normal distribution at 0.01 % significance level. The spatial factor analysis for NO<sub>2</sub> and SO<sub>2</sub> yielded mainly two subregions (Table 1a-b).

Factors	$NO_2$	$NO_2$	$SO_2$	$SO_2$	Deposited dust		
	Summer	Summer Winter Summer		Winter	Summer	Winter	
	half-year	half-year	half-year	half-year	half-year	half-year	
1	2.5	3.4	-	3.4	2.08	3.68	
2	1.6	1.5	-	1.6	1.28	1.22	
3	1.1		-		1.21	1.08	
4			-		1.05		
xpl. var., %	87	81	-	83	56.24	59.79	

Table 1a. The significant eigenvalues and the total percentage of variances explained by the retained and rotated factors, Szeged

However, the *Rule 1*, described in Section 2.1.1, resulted in four subregions for deposited dust in the summer half-year. For each pollutant, summer and winter half-year maps of subregions differ considerably. Most similarity is found between the winter half-year maps of the rotated factor loadings for NO<sub>2</sub> and SO<sub>2</sub>. The summer half-year maps of subregions for

deposited dust are the most confused (Fig. 7.). The eigenvalues and the percentages, explained by the retained and rotated factors explain 55-65 % of the total variance for deposited dust (this value is similar for Szeged and countryside), whilst 60-70 % for NO<sub>2</sub> and SO<sub>2</sub> in countryside, which differs significantly from 80-90 % for those in Szeged (Table 1a-b.) (Makra and Horváth, 1999).

Table 1b. The significant eigenvalues and the total percentage of variances explained by the retained and rotated factors, countryside

Factors	NO <sub>2</sub>	NO <sub>2</sub>	$SO_2$	$SO_2$	Deposi	ted dust
	Summer	Winter	Summer	Winter	Summer	Winter
	half-year	half-year	half-year	half-year	half-year	half-year
1	2.12	3.35	3.13	2.82	3.59	6.38
2	1.59	1.11	1.22	1.47	1.62	1.26
3					1.16	
4					1.01	
Expl. var., %	61.89	74.34	72.49	71.49	61.47	63.64

The method of spatial factor analysis derives the regions from similarities and differences on given time scales. In some cases the regions differ considerably, in other cases they show great similarity. Central parts of the subregions are indicated by the 0.8 factor loading isolines. The regions are perhaps realistic in statistical sense. This means, that they are not direct consequences of the method, itself (Fig. 7.).





a. concentration of deposited dust, winter halfyear, Szeged b. concentration of deposited dust, winter half-year, Csongrád county

Fig. 7a-b. Subareas formed according to the rotated factor loadings when the number of retained factors is > 1.

## 2.2.5. Human biometeorological assessments for air pollution load

Mean (annual) air pollution load (AQSI<sub>1</sub>) became better in 2000 (0.471) and in 1999 (0.448) comparing to its value in 1998 (0.606). This means that the examined busy traffic junction in Szeged can be listed into the air pollution category I. (Mayer, 1995).

A study was made to investigate the difference in daily air pollution load (AQSI<sub>2</sub>) between weekdays and non-weekdays (including Saturdays, Sundays and holidays), using data for the years 1998, 1999 and 2000 at the air quality monitoring station, Szeged. In Hungary, working time is 40 hours per week. It is supposed that the short-term (diurnal) air quality (AQSI<sub>2</sub>) might change during the weekend. The results indicated that AQSI<sub>2</sub> is higher on weekdays and lower during weekends (Table 2.). The air quality is definitely better on holidays in the winter half-year, while role of Saturdays, Sundays and holidays is very similar in the summer half-year. On weekends, air pollution load is lower up to 18 %. Consequently, traffic is supposed to contribute mostly to the change in air pollution load.

Monitoring station Difference of daily averages, % (1)(2)(3) Year -16.22 -8.72 -12.92 Summer half-year -13.52 -13.43 -13.47 Winter half-year -4.64 -18.34 -12.40

Table 2. Difference of air quality stress index (AQSI) values

(1) Difference between Saturday and weekday (Saturday - weekday), %

(2) Difference between holiday and weekday (holiday – weekday), %

(3) Difference between holiday + Saturday and weekday [(holiday + Saturday) - weekday)], %

In order to get an overview of the diurnal air pollution load in Szeged, categorisation of the days were made (see categories in Section 2.1.2.) on the basis of mean diurnal concentrations of air pollutants for the years 1998, 1999 and 2000 (Table 3.) Our results show that number of days with worse air pollution load is higher in the winter half-year (categories II, III. and IV.) comparing to that in the summer half-year. The air quality became substantially better in 1999 as compared with that in 1998, while stagnation can be experienced in 2000. Better air quality is characterised by significantly higher number of days with low air pollution load and, synchronously with this, by considerably lower number of days with extreme pollution load (Table 3.). The winter half-year is more characteristic in occurrences of days with heavier pollution load, than the summer one. This is presented clearly by the number of days, on which daily standards (in  $\mu$ g m<sup>-3</sup>, CO: in mg m<sup>-3</sup>) for air pollutants were exceeded.

Monitoring station	Pollution levels, day											
	Level I.			Level II.		Level III.			Level IV.			
	1998	1999	2000	1998	1999	2000	1998	1999	2000	1998	1999	2000
Year	293	343	335	41	14	8	31	8	24	5	0	2
Summer half-year	173	181	182	5	1	1	5	1	0	1	0	0
Winter half-year	120	162	153	36	13	7	26	7	24	4	0	2

Table 3. Number of days with different pollution levels



Fig. 8. Average diurnal cycle of the pollutants in percentage of their health limits at the monitoring station, Szeged downtown, for the period between 1997-2000.

Daily averages of the pollutants, on the basis of their 30-minute values for the period 1997-2000, were calculated in percentage of the health limits (Fig. 8.). It is concluded that they hardly reach 40 % of the health limits. Even the maximum values, which belong to  $O_3$ , are well below 60 %. This result confirms the above-mentioned consequences of Section 2.2.5, according to which not only the examined busy traffic junction in the neighbourhood of the monitoring station Szeged, but the city itself can be listed into the air pollution category I. (Mayer, 1995).

Hence, diurnal averages of the pollutants between 1997-2000 are far below the 24-hour limit values [CO: 0.45 ppm (limit value: 4.295 ppm); NO<sub>2</sub>: 16.25 ppb (44.34 ppb); (SO<sub>2</sub>: 1.92 ppb (56.19 ppb); O<sub>3</sub>: 20.42 ppb (50 ppb); TSP: 36,72  $\mu$ g m<sup>-3</sup> (100  $\mu$ g m<sup>-3</sup>)].

## **3. CONCLUSIONS**

• Trends of SO<sub>2</sub> emissions show definite decrease both in Szeged and Csongrád county.

• Diurnal and weekly cycles of total suspended particulate are connected with traffic density, as secondary reason, while its annual cycle is influenced by dilution, as cause of intensive vertical exchange in the atmosphere.

• Joint daily behaviour of  $O_3$ -light,  $O_3$ -NO and  $NO_2$ -light pairs show characteristic diurnal courses, which can be explained by their interrelationships.

• The eigenvalues, explained by the retained and rotated factors explain less part of variance for deposited dust than for  $NO_2$  and  $SO_2$ . Spatial factor analysis yielded mainly two subregions for  $NO_2$  and  $SO_2$ , while three-four subregions for deposited dust. Summer half-year maps of factor loadings are more confused than winter half-year ones.

• The air quality stress index for mean (annual) air pollution (AQSI<sub>1</sub>) has definitely decreased in Szeged. Short-term (diurnal) air pollution (AQSI<sub>2</sub>) increased in weekdays and decreased on weekends.

• Daily average concentrations of the pollutants increased in weekdays and decreased during weekends. The concentration of  $O_3$  presented an opposite trend. On weekends, air quality improves better in the winter half-year.

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