ACTA CLIMATOLOGICA ET CHOROLOGICA
Universitatis Szegediensis, Tom. 34-35, 2001, 23-44.

# ANALYSIS OF AIR QUALITY PARAMETERS IN CSONGRÁD COUNTY

L. MAKRA[1], SZ. HORVÁTH[1], A. ZEMPLÉNI[2], V. CSISZÁR[2], ZS. FODRÉ[3], BUCSINÉ I. KAPOCSI[3], G. MOTIKA[4] and Z. SÜMEGHY[1]

[1]*Department of Climatology and Landscape Ecology, University of Szeged, H-6701 Szeged, P.O.B. 653, Hungary; E-mail: makra@geo.u-szeged.hu*
[2]*Department of Probability Theory and Statistics, Eötvös Loránd University, H-1053 Budapest, Kecskemeti u. 10-12, Hungary*
[3]*State Health and Medical Service for Csongrád County; H-6726 Szeged, Derkovits fasor 7-11, Hungary*
[4]*Environmental Protection Inspectorate of Lower-Tisza Region, H-6701 Szeged, P.O.B.1048*

**Összefoglalás** - A tanulmány célja a légszennyező paraméterek időbeli jellemzőinek és statisztikai kölcsönhatásainak vizsgálata Szegeden és Csongrád megyében. Néhány állomás $NO_2$- és $SO_2$-koncentráció idősoraira szélsőérték modelleket illesztünk, s e modellekre alapozva kiszámítjuk a visszatérési időket, azaz a vizsgált két paraméter adott együttes maximális koncentrációit meghaladó értékek jövőbeni legkorábbi együttes bekövetkezéséig terjedő időszakokat. A dolgozatban a vizsgált állomások havi átlagos $NO_2$- és $SO_2$-koncentrációinak, valamint az ülepedő por havi összegeinek idősorait elemezzük. Az adatok - az egyes légszennyező paraméterektől függően - 1985-től állnak rendelkezésre. Az egyes idősorok szerinti trendanalízis eredményei alapján nem tudunk egyértelmű térbeli rendszert kimutatni, mivel egyrészt a szignifikáns trendek periódusai eltérő előjelűek, másrészt az azonos előjelű szignifikáns trendek periódusai eltérők. Alkalmazzuk a kétmintás t-próba egy speciális esetét is, melyet Makra fejlesztett ki. Ez a próba lehetővé teszi annak eldöntését, hogy szignifikáns eltérés van-e két nem független, normális eloszlású minta középértékei között? E próba felhasználásával végzett ún. szakadásvizsgálat szerint az ülepedő por havi összegeinek idősora szignifikáns negatív előjelű szakadást mutat az idősor végén. A vizsgált paraméterek térbeli kapcsolatait faktoranalízis segítségével elemeztük. A célunk az volt, hogy az $NO_2$- és $SO_2$-koncentrációk, valamint az ülepedő por idősorai alapján objektív alrégiókat határolhassunk körül. A légszennyezők arányai a gépjárműforgalom lényeges szerepére utalnak. A forgalom változásai jelentős mértékben hozzájárulnak a levegőminőség napi változásához.

**Summary** - The aim of the study is to determine partly spatial and temporal characteristics, partly statistical interrelationships concerning contaminating parameters at the town of Szeged and in Csongrád county. We also fit extreme-value models to the $NO_2$ and $SO_2$ concentrations observed at some stations and calculate return levels, which are about to be exceeded once in a given period, based on these models. Monthly averages for $NO_2$ and $SO_2$ concentrations and monthly totals for deposited particulates at given sites are analysed in this paper. Data have been available, depending on the pollutants, since at least 1985. Local trend analysis does not show a clear spatial structure among the sites since signs of significant periods are different and significant periods of the same sign are not similar among the sites. A special case of the two-sample t-test developed by Makra is also applied. This test makes it possible to determine whether or not averages of non-independent variables differ significantly. By using this test we made a so called tear analysis, according to which it was found that monthly totals of deposited dust tore considerably with negative signs at the end of the data sets. Spatial relations are analysed by factor analysis with the intention to determine objective subregions by applying data sets of $NO_2$, $SO_2$ and deposited dust. Ratios of pollutants refer definitely to the role of motor vehicle traffic. Variation of traffic contributes mostly to the change in daily air quality.

*Key words:* air pollutants, RIE-network, monitoring station, trend analysis, a special case of the two-sample t-test, extreme value analysis, factor analysis

## INTRODUCTION

Air pollution is one of the greatest environmental problems facing mankind. In many urban areas large concentration of human activities induce considerable amounts of pollutants to be accumulated. As it is well known, the main pollution sources are motor vehicle traffic, which heavily affects air quality in densely urbanized regions, and emissions from building heating systems contribution of which is important in the winter months. Air pollution is harmful to the buildings, technical devices and may cause serious health damage, as well. The nature and importance of air quality problems depend on geographical (climate, local meteorological conditions at the moment, position, relief) and social (existing environmental regulations, urban planning choices) factors. Research of urban air has a wide literature. According to the subject of the papers the authors can make an analysis on characteristics of pollutants (e.g. *Morawska et al.*, 1998), others deal with spatial and temporal variety of those (e.g. *Hastie et al.*, 1996; *Tripathi et al.*, 1996) or investigate statistical interrelationships among variability of pollutants (e.g. *Spicer et al.*, 1996) or examine social policy on regulating emissions (e. g. *Chin*, 1996; *Fang and Chen*, 1999) or study connection of air pollution with meteorological components (e.g. *Kassomenos et al.*, 1995) or evaluate urban air quality using special methods (e.g. *Angius et al.*, 1995) and special air quality indicators (e.g. *Kassomenos et al.*, 1999).

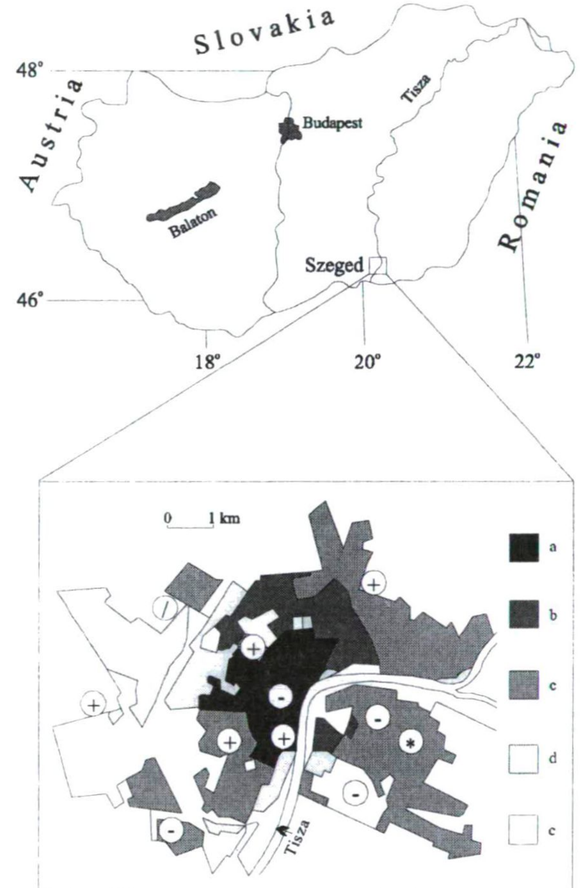


*Fig. 1a* Geographical position of Szeged, Hungary and built-up types of the city (*Unger*, 1997)
(a : centre (2-4-storey buildings); b: housing estates with prefabricated concrete slabs (5-10-storey buildings); c: detached houses (1-2-storey buildings); d: industrial areas; e: green areas; Ø : automatic environmental monitoring station; + : sites of measurements for $SO_2$, $NO_2$ and deposited dust; * : site of measurements for $SO_2$ and $NO_2$ only; - : sites of measurements for deposited dust only; +, *, - : RIE-network)

The aim of the present study, considering the above mentioned classification, is complex: to determine partly spatial and temporal characteristics, partly statistical interrelationships concerning contaminating parameters and to analyze their connection with meteorological elements. The analysis is made on the data of a middle-sized town, Szeged. The analysis is made on the data sets of 9 settlements in Csongrád county (*Fig. 1b*), with special interest to the middle-sized town, Szeged, which is the largest city in the county.

## GEOGRAPHICAL POSITION, METEOROLOGY AND TOPOGRAPHY OF SZEGED AND CSONGRÁD COUNTY

Szeged lies at approximately 20°06'E and 46°15'N near the confluence of the Tisza and Maros rivers. It is the largest town in the south-eastern part of Hungary. The city is flat and low (79 m above sea level), therefore its climate is free from orographical effects (*Fig. 1a*). Consequently its geographical conditions are favourable for the development of an undisturbed urban climate. The number of inhabitants of the city is up to 155,000 and the surface of its built-up area is about 46 km$^2$. The total urban spread extends well beyond the city limits and includes north of the town the largest oil field in Hungary with several oil torches. This oil field is a significant source of $NO_x$ and sulfur dioxide. The power station, located in the western part of the town, and motor vehicle emissions have largely contributed to the nitrogen oxide levels in Szeged.

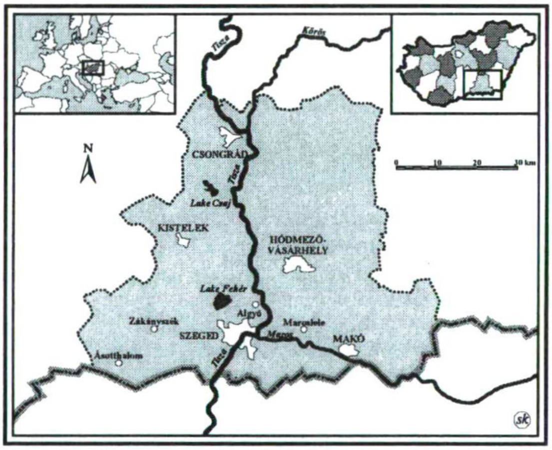


*Fig. 1b* Geographical position of the measuring stations in Csongrád county

Most territory of Hungary (including Csongrád county and Szeged agglomeration) are characterized in climatic classification of Köppen by *Cf* climate (temperate warm climate with quite equal distribution of precipitation amounts) or in that of Trewartha by *D.1* climate (continental climate with a long warm season). Urban air quality basically differs depending on the meteorological elements. The averages for those in Szeged region are as follows. Mean annual temperature is 11.2°C while mean January and July temperatures are -1.2°C and 22.4°C, respectively. As for yearly averages, the annual precipitation is 573 mm, the relative humidity is 71%, the windspeed is 3.2 m s$^{-1}$, and the sunshine duration is 2102 hours.

A detailed climatic classification for Hungary is based on the mean temperature of the growth season ($t_{VS}$) and the aridity index ($H$) (where $H = Q^*/L_vP$ and $Q^*$ is the annual mean net radiation and $L_v$ is the latent heat of vaporization and $P$ is the annual mean precipitation). According to these physical climatological characteristics climate of Szeged is *warm-dry* with $t_{VS} > 17.5$°C and $H > 1.15$.

The basis of the city structure is a boulvard-avenue street system crossed by the River Tisza (*Fig. 1a*). In this way the structure of the city is simple however, following to this system, motor vehicle traffic as well as air pollution are concentrated in the city. The industrial area is located mainly in the north-west part of the town. Thus the prevailing westerly and northerly winds transport the pollutants originating from this area towards the centre of the city.

During the last decades the structure of built-up areas has been significantly modified since huge housing estates were built with prefabricated concrete slabs in the north and east outskirts. These housing estates are found between the centre and the area of detached houses (*Fig. 1a*).

There have been several papers on urban climate of Szeged (e.g. *Unger,* 1997). However, contrary to the fact that there has been operating an environmental monitoring system for more than two decades, investigations on air quality of the city have just been started (*Makra,* 1999).

In Csongrád county (besides Szeged), there have been working altogether 32 RIE-stations. From them 8 stations, distributed almost homogenously, were chosen for further investigation (Hódmezővásárhely, Csongrád, Makó, Kistelek, Ásotthalom, Algyő, Zákányszék and Maroslele) (*Fig. 1b*).

## SAMPLING AND ANALYSIS

An environmental operating system has been working at Szeged since 1985 in the frame of which occasional measurements have been performed by a Regional Immission Examinating (RIE) network of station at various sites of the city for determining concentration of various pollutants (*Table 1*).

The main characteristics of pollutants measured are as follows.

*Sulfur dioxide ($SO_2$)*

There are two main sources of sulfur dioxide emission. The first results from the burning of fuel containing sulfur, such as fuel oil and industrial diesel. The other is from oil refineries and sulfuric acid manufacturing plants. Long-lasting high concentrations of $SO_2$ increases the frequency of respiratory deseases.

*Table 1* Data basis

| *Measurements* | *Pollutant* | *Unit* | *Measurements have been performed since* | *Number of sites* |
|---|---|---|---|---|
| *RIE-network* | $SO_2$ | $\mu g\ m^{-3}$ | 1988 - | 9 |
| | $NO_2$ | $\mu g\ m^{-3}$ | 1988 - | 9 |
| | soot | $\mu g\ m^{-3}$ | in 1992, only | 2 |
| | fine suspended particulates | $\mu g\ m^{-3}$ | 1987 - | 12 |
| | deposited particulates | $g\ m^{-2}$ month | 1985 - | 10 |
| *monitoring station* | CO, NO, $NO_2$, $NO_x$ (= NO + $NO_2$), $O_3$, $SO_2$, fine suspended particulates | $\mu g\ m^{-3}$ | August 1, 1996 | 1 |

*Nitrogen oxides (NO, $NO_2$ and $NO_x$(=NO+$NO_2$))*

Major sources of these pollutants are fuel burning equipments and vehicle exhaust.

*Ozone ($O_3$)*

In tropospheric background air ozone partly comes from photolysis of $NO_x$ and interaction of various organic compounds. As it is known for some conditions the process of ozone formation is controlled almost entirely by $NO_x$ and is largely independent of VOC, while for other conditions ozone production increases with increasing VOC and does not increase (or sometimes even decreases) with increasing $NO_x$. However it has been difficult to determine whether ozone production during specific events is associated with $NO_x$-sensitive chemistry or VOC-sensitive chemistry. For analyzing interrelationships between ozone, $NO_x$ and VOC a model was established (*Sillman,* 1999).

*Carbon monoxide (CO)*

The main source of CO is motor vehicle emission. Others come from incomplete combustion of fuels and cigarette smoke.

*Dust*

Dust, together with suspended particles and smoke, belongs to the group of pollutants named particulate matter (PM). Suspended particles and smoke, which are smaller and lighter, are of greater importance. These reamin in the atmosphere longer and affect a larger area. The effects of dust particles (being larger and heavier) are more localized and they settle quickly. The main sources of particulate matter, in this way that of dust, are motor vehicles, construction activities and fuel burning equipments such as boilers and furnaces.

Samplings for $SO_2$ and $NO_2$ have been performed by AEROMAT OH-601 instruments while analysis of the samples has been occurred by using an automatic analitical instrument named CONTIFLO. Soot and fine suspended particulates were collected on filters by using KS-303 and KS-303 150.10 instruments while deposited particles on plastic pots.

The automatic environmental monitoring station is situated in the downtown, about 15 m away from a busy highway. It measures, besides the pollutants mentioned above, climatic elements (air temperature, humidity, radiation, pressure, precipitation, wind speed and wind direction), as well.

Analysis of occasional measurements performed at various sites of the city is quite difficult because of the unsystematic collection of data. Measurements were performed not every day even at some sites there are considerable lack of data. In order to keep information being available in data, monthly averages of pollutants were calculated if at least ten measurements for a given pollutant were performed in a month. If there were less than ten measurements, the pollutant was taken out of consideration in the given month. By using monthly averages, lack of data was reduced. Even though there are no monthly averages of $SO_2$ and $NO_2$ concentrations between October 1997 and March 1998 at the site Tarján and in October 1997 and between June and December 1998 at the site Ironworks. Data of soot are so much incomplete that it was impossible to make a detailed analysis of them. Deposited particulates have the most complete data series, however each site has some shortage of data and even there were no measurements at the site House of Young Guards between July 1995 and December 1998. Because of absence of data, monthly mean concentrations of $SO_2$ and $NO_2$ (sites: Szeged University Colloidics, Tarján district, Ironworks and Tavasz street) are analyzed between 1995 and 1998. Monthly totals of deposited dust are almost complete for the period between 1985-1998 (ten sites). Incomplete data sets of fine suspended dust makes it impossible to perform a detailed analysis, therefore this parameter was taken out of consideration from further investigations (*Fig. 1a*).

There are also some absence of data at the automatic environmental station (*Table 2*). However pollutants mentioned in *Table 2* are all drawn into the analysis.

*Table 2* Lack of data at the automatic environmental station, in percentage of days of the year

| *pollutant*<br>*year* | *CO* | *NO$_x$* | *O$_3$* | *SO$_2$* | *dust* |
|---|---|---|---|---|---|
| *1997* | 7.7 | 32.3 | 5.8 | 93.4 | 12.1 |
| *1998* | 9.6 | 9.3 | 21.1 | 9.6 | 26.0 |
| *1999* | 0.3 | 3.3 | 1.4 | 0.0 | 25.8 |

## METHOD

### *Trend analysis*

Significancy of linear trends during any subperiod within a data series of given length is checked by Student's t-test, as follows. Let us have the variable of Student distribution,

$$t = (b - \beta) / s_b \tag{1}$$

where $\beta$ - the real (unknown) regression coefficient,

b - empirical regression coefficient, estimated from the finite sample,

$s_b$ - standard deviation of the empirical estimate from the regression coefficient, b.

The zero-hypothesis is that $b = \beta = 0$, i.e. the empirical regression b does not significantly differ from 0. The statistical decision concerning this hypothesis is performed on the basis of the knowledge of the t-distribution (included into tables, in practice). The following analysis of the significance is performed at the 1% significance level, considering that the appropriate degree of freedom is n - 2 , where n is the number of elements in the sample. If the t-value calculated by (1) is higher than the given threshold of the t-distribution, we consider b to be significantly different from 0. Otherwise, it is not. This test has been performed for all possible 3, ..., n years subperiods for the given samples.

### *A special case of the two-sample t-test*

A new statistical test is developed by *Makra* for determining if there is significant difference between expected values of non-independent time series (*Makra et al.*, 2000a).

The developed expression $\dfrac{\overline{M} - \overline{m}}{\sqrt{\dfrac{N-n}{N \cdot n}} \cdot \sigma}$ is a probability variable with N(0;1) distribution.

Now, from the table of the distribution function of the standard normal distribution, it can be determined that $x_p$ to a given $0 < p < 1$ number for which:

$$P\left(\left|\frac{\overline{M} - \overline{m}}{\sqrt{\frac{N-n}{N \cdot n}} \cdot \sigma}\right| > x_p\right) = p \tag{2}$$

If the absolute value of the above probability variable with N(0;1) distribution is higher than $x_p$ then it is said that $\overline{M}$ and $\overline{m}$ differ significantly. The 0-hypothesis, according to which there is no difference between $\overline{M}$ and $\overline{m}$, is realized not more than at the critical p probability.

Significance-tests are carried out at $p = 0.01$ probability level.

### *Extreme value distributions*

The essence of the theory is that if we have a sequence of independent, identically distributed observations, and take their (suitably normalised) maxima, then in most cases it can be approximated by an extreme value distribution. Therefore, if we have a long data series coming from any reasonable distribution (see for example 1 for the exact conditions), then the above-mentioned three parameters can be estimated. The estimation can be based either on the annual (monthly, bi-weekly, etc) maxima or on the highest *k* values (for a summary of the methods, see *Reiss and Thomas,* 1997).

The extreme value distributions form a three-parameter family. Two of them are the shift and scale parameters, which only determine the expected value and the standard deviation of the distribution; while the third one is the so-called „shape" parameter, which has a major effect on the shape of the distribution. This parameter determines if there is finite right or left endpoint of the distribution, as well as the number of existing moments of the distribution. Consequently, if any extreme value distribution is fitted to a given data series, then the abovementioned three parameters should be estimated (*Weissman,* 1977; *Embrechts,* 1997.).

### *Regionalisation by factor analysis*

One of the best methods of studying 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 to 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 standardized variable. Another vital stage in this analysis is the so-called rotation of the axes (factors). This process achieves a discrimination among the loadings which makes the rotated axes easier to interpret. In this analysis the *Orthogonal Varimax Rotation* has been applied, which keeps the factors uncorrelated. In general, there is no guarantee that the evaluated factors represent dynamically existing entities, but, as with any statistical tool, it is important to determine whether the results have any physical meaning.

## RESULTS AND DISCUSSION

On the basis of the local trend analysis our findings are as follows. Linear trends of monthly amounts of deposited dust (μg $m^{-2}$) and that of $NO_2$ concentrations show significant trends both for relatively short (7-23 months) and quite long (102-137 months) sequences. It is also remarkable that the number of significant trends is at some stations 1 or 2, while at other stations 4 or 5. For the longer sequences, the interannual variance is too high to allow significance of monotonous linear changes in the local series. In the table, sequence of significant periods is set in diachronic order. If comparing signs of significant periods, one can establish that they are completely different at the sites. Significant periods of the same sign are not much similar among the sites. This means that short sequences are yielded rather by random interannual fluctuations, than by the long-term trends (*Table 3a-b*).

*Table 3a* Subperiods with significant trends of deposited dust in 1985-1998; year, month (m)

| ***Teachers' Training College*** | ***Tarján district*** | ***Iron-works*** | ***Esperanto street*** | ***Bécsi Boulevard*** | ***State Service for Health Control, New Szeged*** | ***Nurses' Training School, New Szeged*** | ***Széchenyi square*** | ***Mátyás square*** |
|---|---|---|---|---|---|---|---|---|
| 1986.07.-<br>1994.02.<br>102 m (+) | 1985.03.-<br>1996.07.<br>137 m (-) | 1987.12-<br>1990.06.<br>51 m (+) | 1989.11.-<br>1998.09.<br>106 m (+) | 1985.01.-<br>1998.03.<br>159 m (-) | 1985:11.-<br>1987.08.<br>22 m (-) | 1986.12.-<br>1988.06.<br>19 m (+) | 1986.03.-<br>1989.03.<br>37 m (-) | 1985.01.-<br>1988.07.<br>43 m (-) |
| 1988.02.-<br>1998.05.<br>124 m (-) | 1989.09.-<br>1998.08:<br>108 m (+) | 1988.04.-<br>1992.10.<br>55 m (-) | | 1989.12.-<br>1992.04.<br>59 m (+) | 1994.04-<br>1995.01.<br>10 m (-) | 1989. 04.-<br>1994.07.<br>64 m (+) | 1990.09.-<br>1992.09.<br>25 m (+) | |
| | | 1991.09.-<br>1995.12.<br>52 m (+) | | 1992.10.-<br>1994.10.<br>25 m (+) | 1994.09.-<br>1998.01.<br>41 m (+) | 1993.06.-<br>1998.12.<br>67 m (-) | 1994.03.-<br>1995.03.<br>13 m (-) | |
| | | 1995.08.-<br>1997.02.<br>19 m (-) | | 1992.12.-<br>1993.07.<br>8 m (+) | | | 1997.02.-<br>1997.08.<br>7 m (-) | |
| | | 1996.02.-<br>1997.12.<br>23 m (+) | | 1996.01.-<br>1996.08.<br>8 m (+) | | | | |

*Table 3b* Subperiods with significant trends of $NO_2$ concentrations in 1995-1998; year, month (m)

| ***Szeged University, Colloidics*** | ***Tarján district*** | ***Ironworks*** | ***Esperanto street*** | ***Bécsi Boulevard*** | ***House of Parties*** |
|---|---|---|---|---|---|
| 1991.01.-<br>1992.06.<br>18 m (-) | 1991.06.-<br>1991.12.<br>7 m (-) | 1991.01.-<br>1993.11.<br>36 m (+) | 1991.02.-<br>1991.11.<br>10 m (-) | 1991.01.-<br>1993.11.<br>35 m (-) | 1991.10.-<br>1993.03.<br>18 m (+) |
| 1991.07.-<br>1993.12.<br>30 m (+) | 1992.10.-<br>1993.02.<br>5 m (+) | | 1991.07.-<br>1993.04.<br>22 m (+) | | 1993.02.-<br>1993.06.<br>5 m (-) |
| | | | 1992.02.-<br>1992.05.<br>4 m (-) | | |
| | | | 1993.01.-<br>1993.07.<br>7 m (-) | | |

*Table 3c* Subperiods with significantly different averages of deposited dust from that of the full data set in 1985-1998; year, month (m)

| ***Teachers' Training College*** | ***Tarján district*** | ***Iron-works*** | ***Esperanto street*** | ***Bécsi Boulevard*** | ***State Service for Health Control, New Szeged*** | ***Nurses' Training School, New Szeged*** | ***Széchenyi square*** | ***Mátyás square*** |
|---|---|---|---|---|---|---|---|---|
| 1985.03.-<br>1996.11.<br>141 m (+) | 1985.03.-<br>1986.07.<br>17 m (+) | 1986.04.-<br>1990.04.<br>49 m (+) | 1985.03.-<br>1985.11.<br>9 m (+) | 1985.02.-<br>1989.10.<br>57 m (+) | 1992.06.-<br>1993.02.<br>9 m (+) | 1985.03.-<br>1995.05.<br>123 m (+) | 1988.03.-<br>1989.04.<br>14 m (+) | 1988.09.-<br>1989.11.<br>15 m (+) |
| 1990.11.-<br>1998.11.<br>97 m (-) | 1986.07.-<br>1997.03.<br>129 m (-) | 1989.11.-<br>1997.04.<br>91 m (-) | 1986.01.-<br>1997.01.<br>133 m (-) | 1989.11.-<br>1998.03.<br>101 m (-) | | 1989.04.-<br>1998.12.<br>117 m (-) | 1989.06.-<br>1996.10.<br>89 m (-) | 1989.02.-<br>1992.08.<br>43 m (+) |
| | 1997.07.-<br>1998.07.<br>13 m (+) | | 1989.02.-<br>1989.08.<br>7 m (+) | | | | 1997.01.-<br>1997.06. 6 m (+) | 1998.02.-<br>1998.06.<br>5 m (+) |
| | | | 1997.02.-<br>1998.08.<br>19 m (+) | | | | | |

*Table 3d* Subperiods with significantly different averages of $NO_2$ concentrations from that of the full data set in 1995-1998; year, month (m)

| ***Szeged University, Colloidics*** | ***Tarján district*** | ***Ironworks*** | ***Esperanto street*** | ***Bécsi Boulevard*** | ***House of Parties*** |
|---|---|---|---|---|---|
| 1991.01.-<br>1991.05.<br>5 m (+) | 1991.04.-<br>1991.09.<br>6 m (+) | 1991.01.-<br>1992.11.<br>23 m (-) | 1992.09.-<br>1993.05.<br>9 m (+) | 1991.01.-<br>1992.03.<br>15 m (+) | 1991.05.-<br>1992.05.<br>13 m (-) |
| 1991.04.-<br>1993.10.<br>31 m (-) | | 1992.08.-<br>1993.12.<br>17 m (+) | | 1991.04.-<br>1993.12.<br>33 m (-) | 1992.06.-<br>1993.04.<br>11 m (+) |

As for the application of the special case of the two-sample t-test, subperiods were identified averages of which are significantly different from the mean of the data series examined. Concentrations of pollutants in these subperiods were much higher or lower than that. The point of the method is to determine the start and the termination of the significant periods. This is done for date for which the significance is still valid, irrespectively to that, which years would yield the strongest difference (i. e. among the significant ones). This search has been performed by a special case of two-sample t-test, applied to detect differences between averages of non-independent data sets (*Makra and Horváth,* 1999; *Makra et al.,* 2000b). The significance tests have been performed at the 1% significance level (*Table 3c-d*). In the table, sequence of significant periods is defined in diachronic order.

The following results can be established. In the data sets of deposited dust, the signs of the most long-term periods (91-133 months) are negative which refer to the fact that at seven sites from the nine the amount of deposited dust decreased significantly. The experienced similarity among the long-term anomalies of identical sign at the sites suggest that these long sequences are already yielded the long-term trends. Significant subperiods in the data sets of $NO_2$ concentrations do not show a clear tendency. At some stations they have positive sign while at other ones, even for the same period, they are negative.

Since not only one measuring station is working at the examined settlements, data maximum of all stations was considered for a given settlement every day. $NO_2$ and $SO_2$ data series for the examined settlements, received in this way, are shown in *Fig. 2a-i* and *Fig. 3a-i*. One can see that these data are incomplete and comparison between data series of the examined settlements promises to be very difficult, since periods with lack of data are different. At some cases there might be even incorrect values in the data series (very high values).

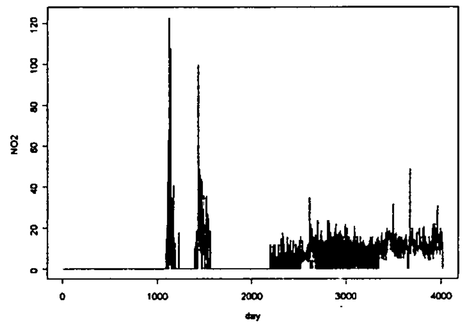


*Fig. 2a* $NO_2$ data series for Szeged, received by taking daily maxima of all stations at Szeged, µg $m^{-3}$

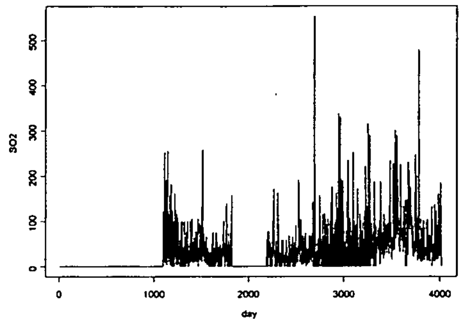


*Fig. 3a* $SO_2$ data series for Szeged, received by taking daily maxima of all stations at Szeged, µg $m^{-3}$

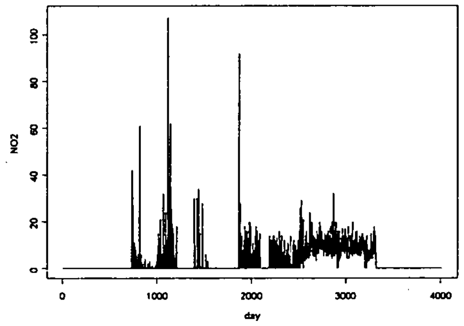


*Fig. 2b* $NO_2$ data series for Hódmezővásárhely, received by taking daily maxima of all stations at Hódmezővásárhely, µg $m^{-3}$

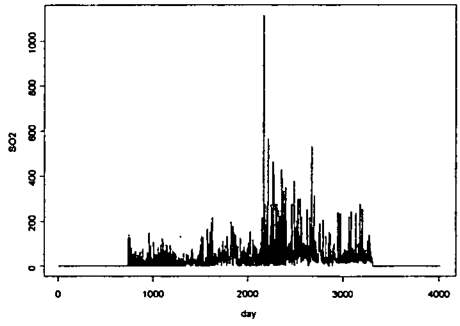


*Fig. 3b* $SO_2$ data series for Hódmezővásárhely, received by taking daily maxima of all stations at Hódmezővásárhely, µg $m^{-3}$

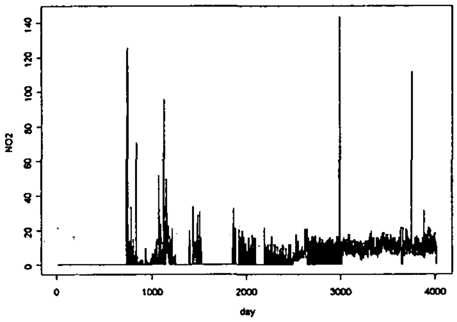


*Fig. 2c* $NO_2$ data series for Csongrád, received by taking daily maxima of all stations at Csongrád, µg $m^{-3}$

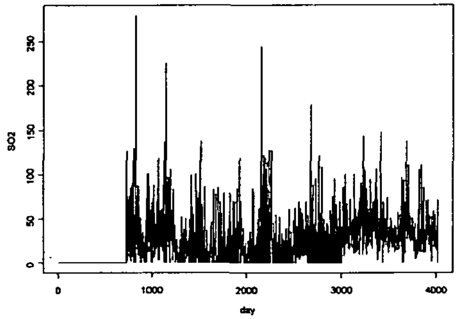


*Fig. 3c* $SO_2$ data series for Csongrád, received by taking daily maxima of all stations at Csongrád, µg $m^{-3}$

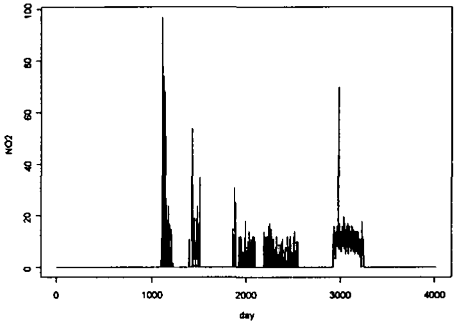


*Fig. 2d* $NO_2$ data series for Makó, received by taking daily maxima of all stations at Makó, µg $m^{-3}$

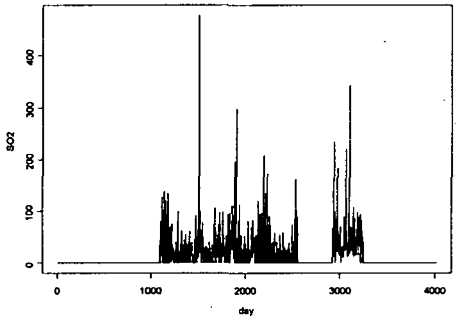


*Fig. 3d* $SO_2$ data series for Makó, received by taking daily maxima of all stations at Makó, µg $m^{-3}$

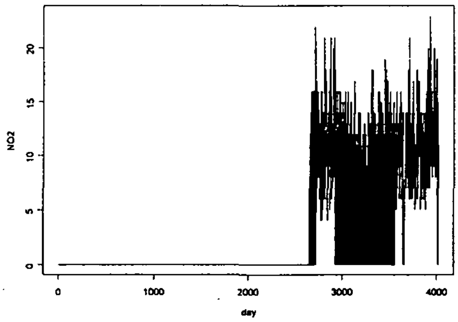


*Fig. 2e* $NO_2$ data series for Kistelek, received by taking daily maxima of all stations at Kistelek, µg $m^{-3}$

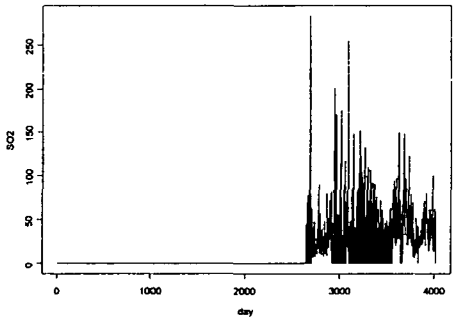


*Fig. 3e* $SO_2$ data series for Kistelek, received by taking daily maxima of all stations at Kistelek, µg $m^{-3}$

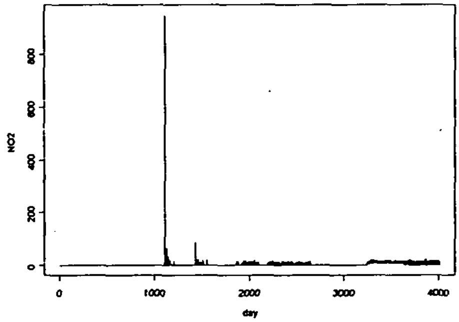


*Fig. 2f* $NO_2$ data series for Ásotthalom, received by taking daily maxima of all stations at Ásotthalom, µg $m^{-3}$

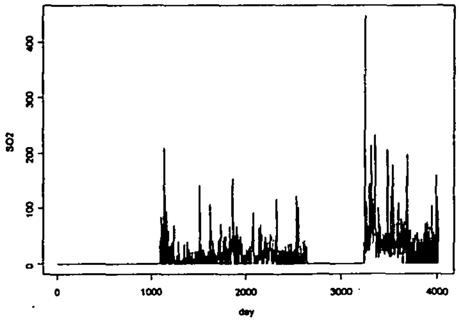


*Fig. 3f* $SO_2$ data series for Ásotthalom, received by taking daily maxima of all stations at Ásotthalom, µg $m^{-3}$

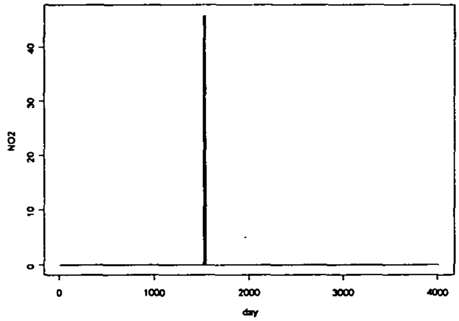


*Fig. 2g* $NO_2$ data series for Algyő, received by taking daily maxima of all stations at Algyő, µg $m^{-3}$

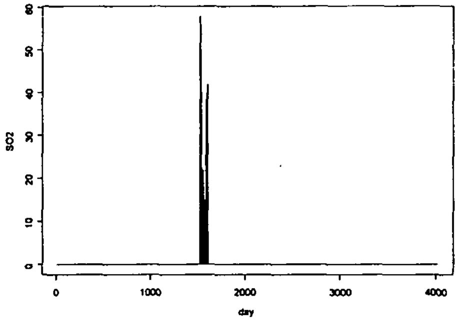


*Fig. 3g* $SO_2$ data series for Algyő, received by taking daily maxima of all stations at Algyő, µg $m^{-3}$

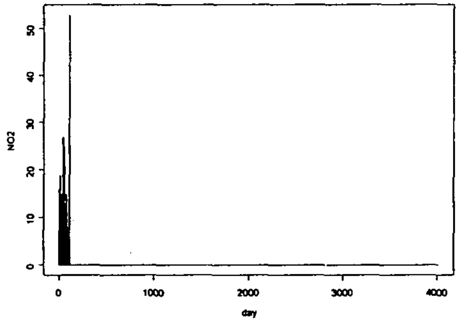


*Fig. 2h* $NO_2$ data series for Zákányszék, received by taking daily maxima of all stations at Zákányszék, µg $m^{-3}$

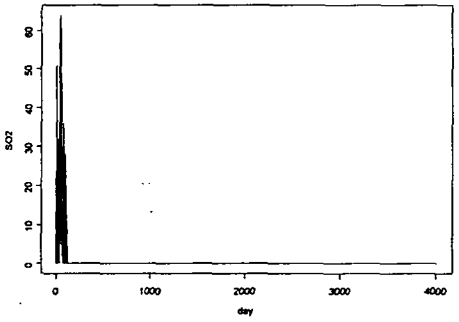


*Fig. 3h* $SO_2$ data series for Zákányszék, received by taking daily maxima of all stations at Zákányszék, µg $m^{-3}$

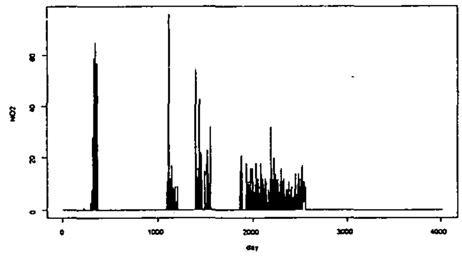


*Fig. 2i* $NO_2$ data series for Maroslele, received by taking daily maxima of all stations at Maroslele, μg m$^{-3}$

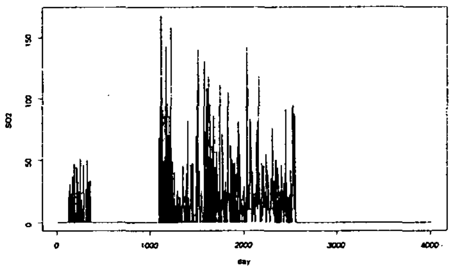


*Fig. 3i* $SO_2$ data series for Maroslele, received by taking daily maxima of all stations at Maroslele, μg m$^{-3}$

Afterwards, bi-weekly maxima of the abovementioned data series were calculated. In this way, the data series were contracted both in space and time. This procedure was important to perform, in order to eliminate lack of data. Namely, $NO_2$ was measured one day and $SO_2$ the other day at most of the measuring stations. However, long enough and unbroken data series were not received even in this way. Consequently, data series being at disposal, were simply pushed together (*Fig. 4a-g, Fig. 5a-g*), implicitly assuming the stationarity of the series.

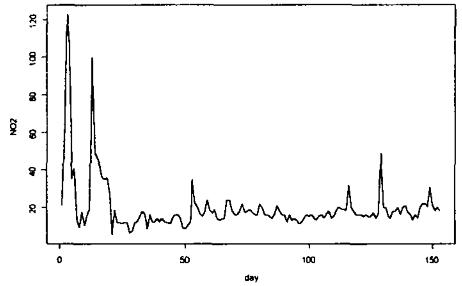


*Fig. 4a* $NO_2$ data series for Szeged; bi-weekly unbroken maxima derived from *Fig. 2a*, μg m$^{-3}$

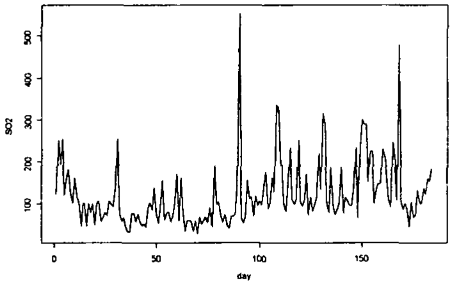


*Fig. 5a* $SO_2$ data series for Szeged; bi-weekly unbroken maxima derived from *Fig. 3a*, μg m$^{-3}$

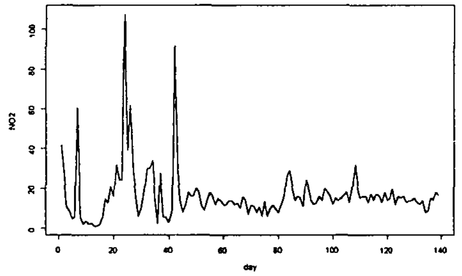


*Fig. 4b* $NO_2$ data series for Hódmezővásárhely; bi-weekly unbroken maxima derived from *Fig. 2b*, μg m$^{-3}$

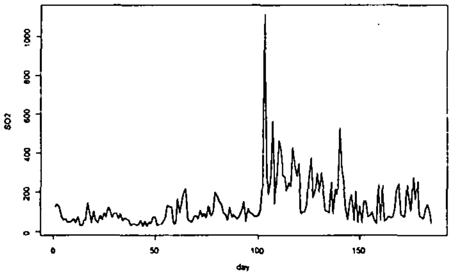


*Fig. 5b* $SO_2$ data series for Hódmezővásárhely; bi-weekly unbroken maxima derived from *Fig. 3b*, μg m$^{-3}$

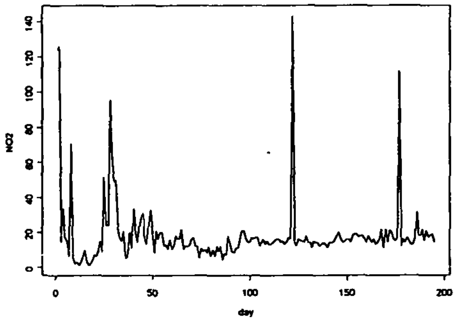


*Fig. 4c* $NO_2$ data series for Csongrád; bi-weekly unbroken maxima derived from *Fig. 2c*, μg m$^{-3}$

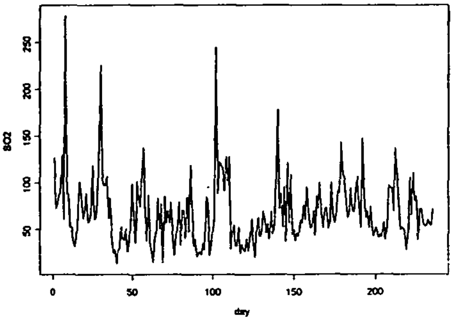


*Fig. 5c* $SO_2$ data series for Csongrád; bi-weekly unbroken maxima derived from *Fig. 3c*, μg m$^{-3}$

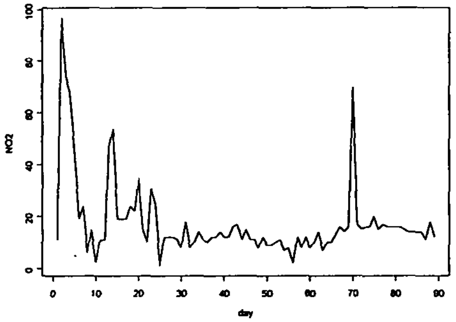


*Fig. 4d* $NO_2$ data series for Makó; bi-weekly unbroken maxima derived from *Fig. 2d*, μg m$^{-3}$

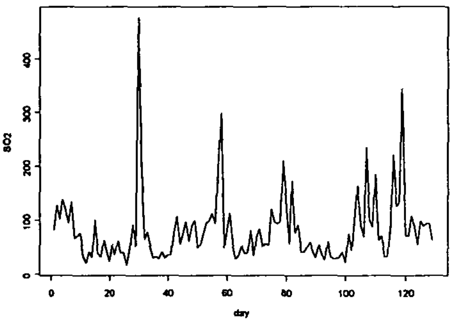


*Fig. 5d* $SO_2$ data series for Makó; bi-weekly unbroken maxima derived from *Fig. 3d*, μg m$^{-3}$

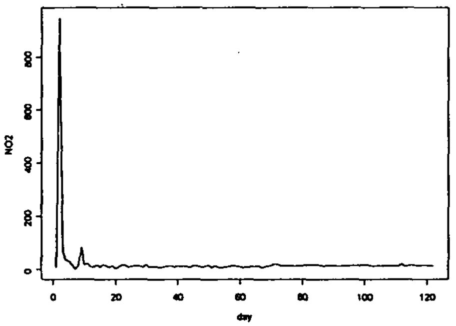


*Fig. 4f* $NO_2$ data series for Ásotthalom; bi-weekly unbroken maxima derived from *Fig. 2f*, μg m$^{-3}$

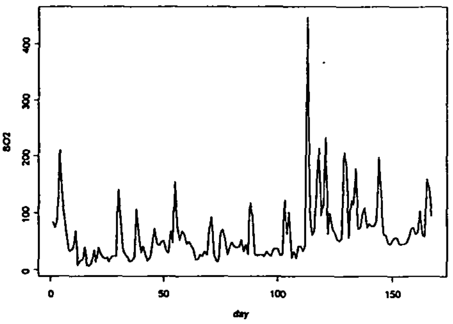


*Fig. 5f* $SO_2$ data series for Ásotthalom; bi-weekly unbroken maxima derived from *Fig. 3f*, μg m$^{-3}$

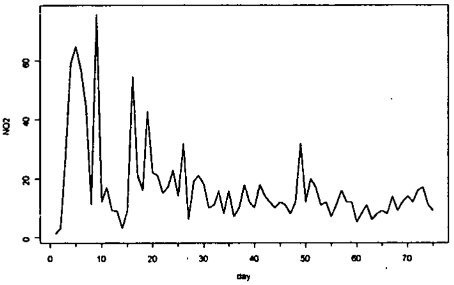


*Fig. 4g* $NO_2$ data series for Maroslele; bi-weekly unbroken maxima derived from *Fig. 2i*, μg $m^{-3}$

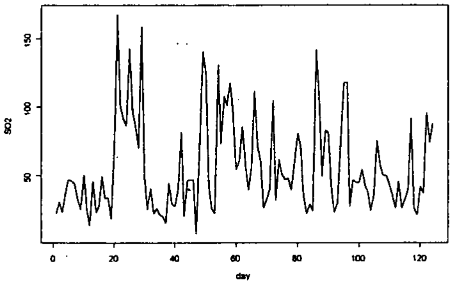


*Fig. 5g* $SO_2$ data series for Maroslele; bi-weekly unbroken maxima derived from *Fig. 3i*, μg $m^{-3}$

After that, autocorrelations of bi-weekly unbroken maxima for Szeged (*Fig. 6a-b*) and Maroslele (*Fig. 7a-b*) were analysed. It is evident that even these data series can not be considered totally independent.

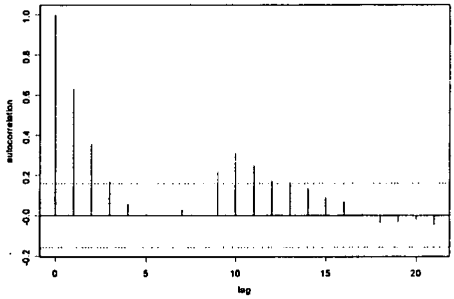


*Fig. 6a* Autocorrelations of the bi-weekly unbroken maxima for Szeged, $NO_2$, μg $m^{-3}$. (Confidence interval is 95%.)

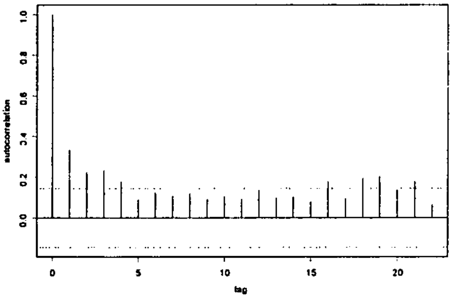


*Fig. 6b* Autocorrelations of the bi-weekly unbroken maxima for Szeged, $SO_2$, μg $m^{-3}$. (Confidence interval is 95%.)

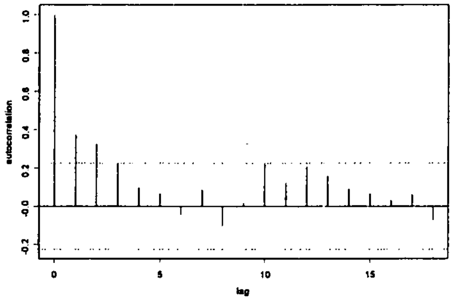


*Fig. 7a* Autocorrelations of the bi-weekly unbroken maxima for Maroslele, $NO_2$, μg $m^{-3}$. (Confidence interval is 95%.)

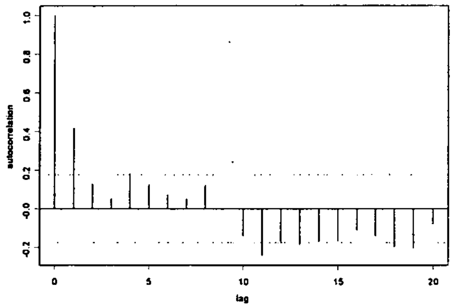


*Fig. 7b* Autocorrelations of the bi-weekly unbroken maxima for Maroslele, $SO_2$, μg $m^{-3}$. (Confidence interval is 95%.)

Further on, considering the data series of the bi-weekly unbroken maxima independent with the same distribution, extreme value distributions were fitted to them, by the statistical procedures described in (*Reiss and Thomas,* 1997).

Extreme value distributions were fitted to both $NO_2$ and $SO_2$ data series of Szeged and 5 further settlements in Csongrád county.

As a general experience, this family of distributions was a poor fit to $NO_2$ data series (*Fig. 8a*), while it was a good fit to those of $SO_2$ (*Fig. 8b*).

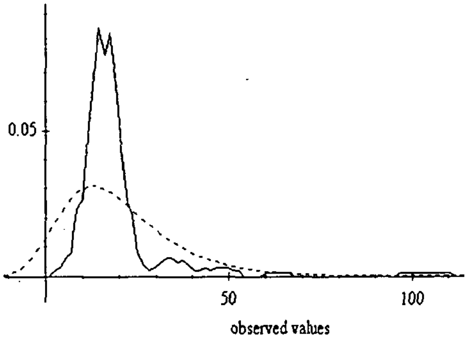


*Fig. 8a* Smoothed empirical density function of the bi-weekly maxima (unbroken line) and density function of the extreme value distribution fitted to that (dotted line), Szeged, $NO_2$. (The fit is poor, cause of which might be the irregular pattern of very high values of the data series.)

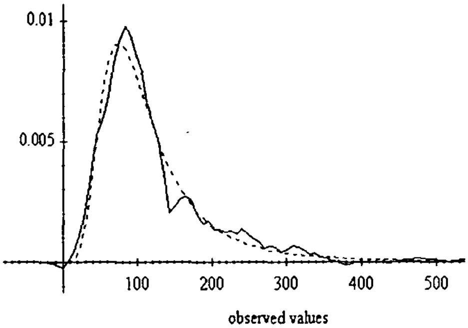


*Fig. 8b* Smoothed empirical density function of the bi-weekly maxima (unbroken line) and density function of the extreme value distribution fitted to that (dotted line), Szeged, $SO_2$. (The fit is good.)

The shape parameter of the extreme value distribution can change depending on how many extreme values of the data series are used (*Fig. 9*). From *Fig. 9* it can be seen that the estimation is jumping upside down and that the figure has no any stable horizontal part. Consequently, no any reliable estimations of the return levels can be taken for the $NO_2$ series. (Return level, corresponding to a given time period is the value, which is expected to be exceeded in the given time period only once.)

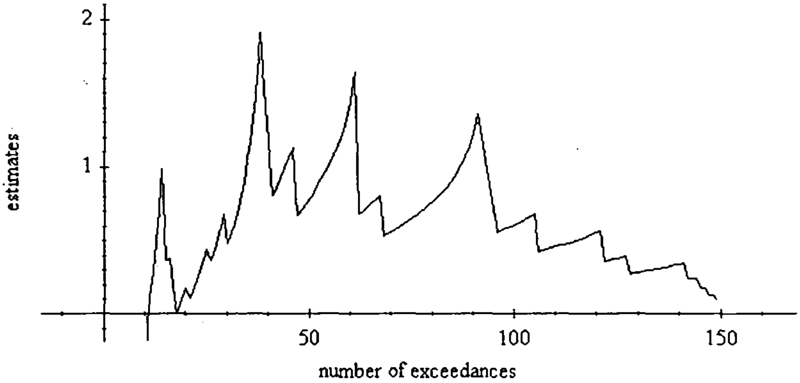


*Fig. 9* Estimation of the shape parameter of the extreme value distribution, depending on the number of extreme values are used for the estimation, Szeged, $NO_2$

As it has already been reported, fitting was unacceptable for $NO_2$ data series at each examined settlements (*Fig. 9*). Findings on unreliability of estimations are illustrated by the following example. At Csongrád station there were 190 values at disposal. First, extreme value distribution was fitted to these starting data, then the data set of maxima coming from the consecutive four-element blocks were formed. In this way, 47 values were only obtained; however, it was supposed that a better fitting would be received for them. Nevertheless, it was not the case; the two estimations differed very much, which is represented in *Table 4*. Positive shape parameters correspond to distributions with infinite right-endpoint. The larger the value of gamma, the heavier the tail of the distribution, for example the moments are finite up to $1/\gamma$.

*Table 4* Estimators for the shape parameter and some return levels, Csongrád, $NO_2$

| ***Data series*** | ***Gamma*** | ***Return levels to the periods*** | | |
|---|---|---|---|---|
| | | ***2 years*** | ***4 years*** | ***10 years*** |
| original | 0.19 | 52.6 | 63.6 | 80.6 |
| maxima of four-element blocks | 0.70 | 133.2 | 221.1 | 425.5 |

Contrary to the results for $NO_2$, extreme value distributions fit very well to smoothed empirical density functions of $SO_2$ data series (*Fig. 8a-b*). In the latter case, estimating parameters and return periods in two different ways, very similar results were received. E.g. for Csongrád station, on the basis of 230 starting observations, results are as follows (*Table 5*).

*Table 5* Estimators for the shape parameter and some return levels, Csongrád, $SO_2$

| ***Data series*** | ***Gamma*** | ***Return levels to the periods*** | | |
|---|---|---|---|---|
| | | ***2 years*** | ***4 years*** | ***10 years*** |
| original | 0.08 | 165.9 | 190.1 | 224.0 |
| maxima of four-element blocks | 0.10 | 165.9 | 195.8 | 238.0 |

Estimations for $SO_2$ data series at the examined 6 stations are summarized in *Table 6*. Estimations for Hódmezővásárhely seem to be too high.

*Table 6* Estimators for the shape parameters and some return levels, $SO_2$

| ***Settlement*** | ***Number of data*** | ***Gamma*** | ***Return levels to the periods*** | | |
|---|---|---|---|---|---|
| | | | ***2 years*** | ***4 years*** | ***10 years*** |
| *Szeged* | 180 | 0.25 | 364.1 | 449.9 | 588.8 |
| *Maroslele* | 120 | 0.23 | 174.0 | 213.9 | 277.5 |
| *Makó* | 120 | 0.35 | 290.9 | 379.2 | 533.5 |
| *Hódmezővásárhely* | 180 | 0.46 | 594.8 | 829.8 | 1280.4 |
| *Csongrád* | 230 | 0.08 | 165.9 | 190.1 | 224.0 |
| *Ásotthalom* | 160 | 0.33 | 237.1 | 307.5 | 428.3 |

Factor analysis defined different numbers of subareas for the concentration of $NO_2$, $SO_2$ and deposited dust in the total, summer half-year and winter half-year data sets. (For the concentration of $SO_2$ only the winter half-year data set was submitted to factor analysis since the other two ones were incomplete.) The eigenvalues and the percentages explained by the retained and rotated factors are shown in *Table 7*. It is found that the retained factors explain 71-87% of the total variance exhibited by all initial variables. The number of retained factors varies between 2 and 4. Two subregions are found both for $SO_2$ and $NO_2$ concentrations in the winter half-year, locations of which are very similar. The map of the rotated factor loadings for $NO_2$ concentrations in the summer half-year shows three subregions which are different from those in the winter half-year. Maps for deposited dust are totally different from those for $NO_2$ and $SO_2$ concentrations. On the maps for deposited dust the analysis yields four subregions in the summer half-year and three in the winter half-year, even these last maps differ substantially from each other (*Fig. 10a-g*).

The method of 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 (*Fig. 10a-g*). 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.

*Table 7* The significant eigenvalues and the total percentage of variance explained by the retained and rotated factors

| ***Pollutant*** | ***$SO_2$*** | | | ***$NO_2$*** | | | ***Deposited dust*** | | |
|---|---|---|---|---|---|---|---|---|---|
| ***Eigen-values*** | ***Year*** | ***Summer half-year*** | ***Winter half-year*** | ***Year*** | ***Summer half-year*** | ***Winter half-year*** | ***Year*** | ***Summer half-year*** | ***Winter half-year*** |
| 1 | - | - | 3.4 | 2.9 | 2.5 | 3.4 | 2.8 | 1.8 | 4.1 |
| 2 | - | - | 1.6 | 1.3 | 1.6 | 1.5 | 1.2 | 1.4 | 1.2 |
| 3 | - | - | | | 1.1 | | 1.0 | 1.2 | 1.0 |
| 4 | - | - | | | | | | 1.1 | |
| expl. var.,% | - | - | 83 | 71 | 87 | 81 | 45 | 61 | 60 |

Based on the annual, summer and winter half-year average ratios of various pollutants and surrounding pollution sources the characteristics of the automatic environmental monitoring station can be summarized as follows (*Table 8*). The station is located near to a highway. Consequently CO averages are high for the whole year and the summer and winter half-years. As $SO_2$/CO ratio is far less than 1, it shows that there are no factories near to the station discharging $SO_2$. Whereas in the surroundings of the station there are no typical industrial areas, the $NO_x$/CO ratio is low. Since the NO/$NO_2$ ratio is no greater than 1, it means that the station is without being significantly influenced by pollution sources from traffic. Small PM(μg $m^{-3}$)/CO(ppdm) ratio (far less than 1) indicates that PM sources are highly related to vehicles' activities (*Fang and Chen,* 1999). Considering the results, there are no significant differences except for the NO/$NO_2$ ratio in 1998 and the CO concentration in 1997 and 1998, respectively. In the winter half-year the values of the above mentioned parameters are double than those in the summer half-year.

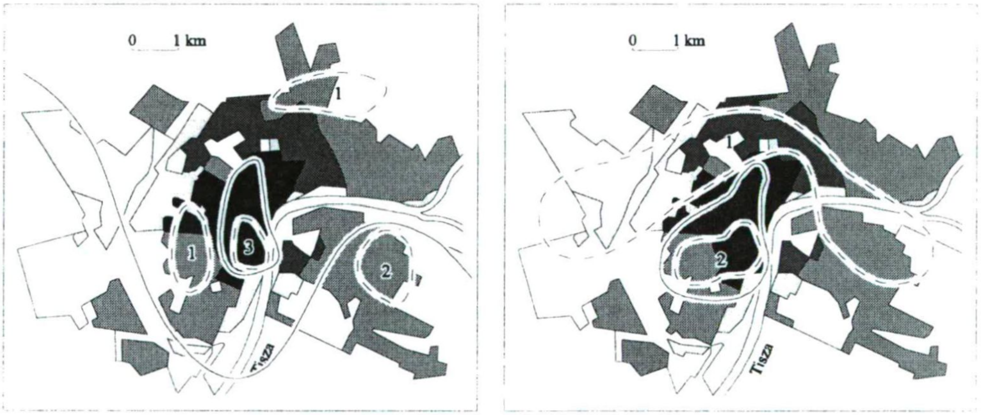
A study was conducted to investigate the difference in concentration of pollutants between weekday and non-weekday (including Saturday, Sunday and holiday) by using the 1997 and 1998 data from the automatic environmental station. In Hungary working time is 44 hours per week. It is speculated that the air quality might change during the weekend. It was found that the variation of traffic contributed mostly to the change in daily air quality. The results coming from the automatic environmental station also indicated that the daily average concentration of $SO_2$, CO, $NO_x$, NO, $NO_2$ and PM all increased in weekdays and decreased during the weekend (*Table 9a-c*). However, the $O_3$ daily average exhibited the opposite trend, it increased on Saturdays, Sundays and holidays but decreased on weekdays. The findings regarding $O_3$ level might indicate that the monitoring station is located near to a highway. Therefore the reaction between $O_3$ and NO is fast. $O_3$ increases as NO decreases on Saturdays, Sundays and holidays and vice versa (*Fang and Chen,* 1999).

*Table 8* Annual, summer- and winter half-year averages of $SO_2$/CO, $NO_x$/CO, NO/$NO_2$, PM/CO ratios and those of CO in 1997, 1998 and 1999* at the automatic station

| ***Period*** | ***$SO_2$/ CO*** | | | ***$NO_x$/ CO*** | | | ***NO / $NO_2$*** | | |
|---|---|---|---|---|---|---|---|---|---|
| | ***1997*** | ***1998*** | ***1999*** | ***1997*** | ***1998*** | ***1999*** | ***1997*** | ***1998*** | ***1999*** |
| *year* | 0.022 | 0.010 | 0.011 | 0.077 | 0.107 | 0.093 | 0.385 | 0.517 | - |
| *Apr-Sept* | - | 0.008 | 0.008 | 0.092 | 0.123 | 0.088 | 0.311 | 0.325 | - |
| *Oct-March* | 0.016 | 0.010 | 0.013 | 0.081 | 0.103 | 0.097 | 0.453 | 0.669 | - |

| ***Period*** | ***PM / CO*** | | | ***CO*** | | |
|---|---|---|---|---|---|---|
| | ***1997*** | ***1998*** | ***1999*** | ***1997*** | ***1998*** | ***1999*** |
| *year* | 0.071 | 0.076 | 0.083 | 5188 | 5216 | 4188 |
| *Apr-Sept* | 0.098 | 0.099 | 0.100 | 3214 | 3462 | 3087 |
| *Oct-March* | 0.061 | 0.063 | 0.072 | 6900 | 6660 | 5288 |

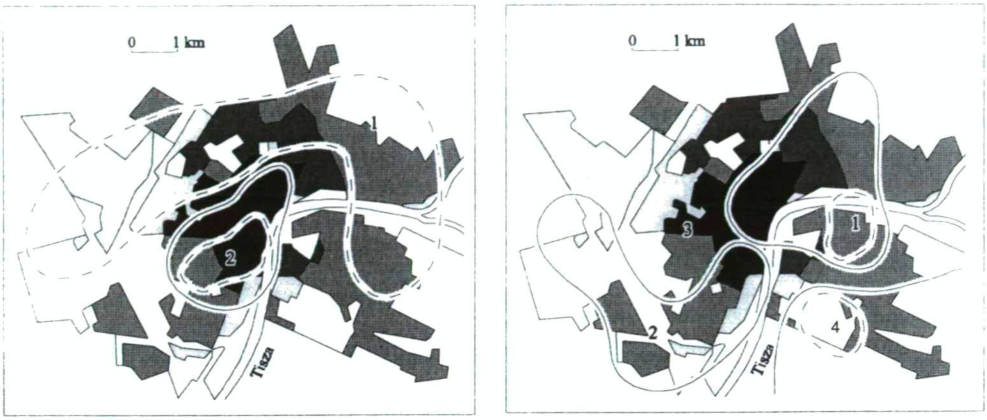
*$SO_2$, $NO_x$ (NO, $NO_2$), PM in µg $m^{-3}$, CO in ppdm



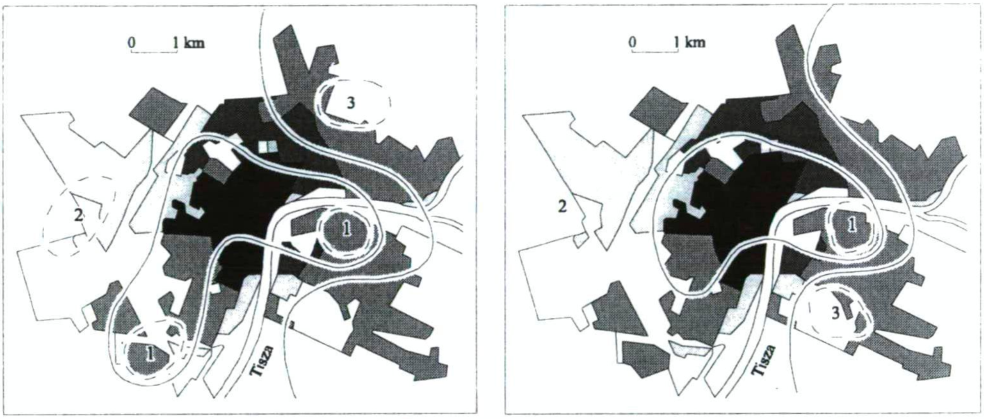
*a.* $NO_2$ concentration, summer half-year

*b.* $NO_2$ concentration, winter half-year

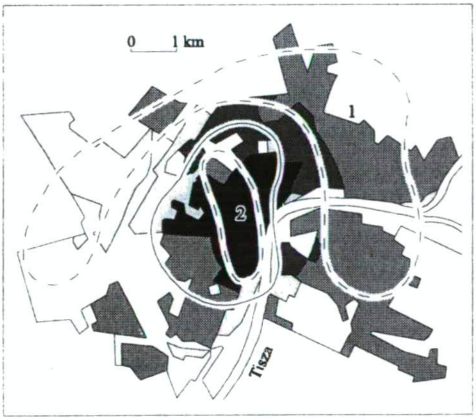
*Fig. 10a-g* Subareas formed according to the rotated factor loadings when the number of retained factors is > 1. Isopleths of loadings 0.8 or higher are indicated.



*c*. $NO_2$ concentration, year *d*. concentration of deposited dust, summer half-year



*e*. concentration of deposited dust, winter half-year *f*. concentration of deposited dust, year



*g*. $SO_2$ concentration, winter half-year

*Fig. 10a-g* (continued)

*Table 9a* Air quality difference between Saturday and weekday (Saturday – weekday),%

| ***Automatic station*** | ***Difference of daily average*** | | | | |
|---|---|---|---|---|---|
| *pollutant* | CO | $NO_x$ | $O_3$ | $SO_2$ | dust |
| *year* | -12.5 | -22.6 | 7.2 | -12.6 | -9.4 |
| *summer half-year* | -9.9 | -25.1 | 6.2 | -19.6 | -9.2 |
| *winter half-year* | -13.5 | -16.9 | 6.4 | -3.4 | -8.6 |

*Table 9b* Air quality difference between holiday and weekday (holiday – weekday),%

| ***Automatic station*** | ***Difference of daily average*** | | | | |
|---|---|---|---|---|---|
| *pollutant* | CO | $NO_x$ | $O_3$ | $SO_2$ | dust |
| *year* | -17.1 | -41.0 | 10.4 | -25.7 | -17.9 |
| *summer half-year* | -14.2 | -31.5 | 9.1 | -15.6 | -17.0 |
| *winter half-year* | -21.8 | -51.8 | 12.9 | -36.6 | -19.6 |

*Table 9c* Air quality difference between holiday+Saturday and weekday [(holiday+Saturday) – weekday)],%

| ***Automatic station*** | ***Difference of daily average*** | | | | |
|---|---|---|---|---|---|
| *pollutant* | CO | $NO_x$ | $O_3$ | $SO_2$ | dust |
| *year* | -15.0 | -32.6 | 8.9 | -19.8 | -14.1 |
| *summer half-year* | -12.1 | -28.5 | 7.8 | -17.5 | -13.3 |
| *winter half-year* | -18.2 | -36.3 | 10.0 | -22.2 | -14.9 |

**Acknowledgement** – The authors are indebted to L. Lukácsovics for fruitful discussions. The authors thank L. Csikász for his contribution with digital mapping. This research was supported by the grants from the Ministry of Education (FKFP-0429/1997 and FKFP-0001/2000) and OTKA (T-34765).

## REFERENCES

*Angius, S.P., Angelino, E., Castrofino, G., Gianelle, V., Tamponi, M. and Tebaldi, G.*, 1995: Evaluation of the effects of traffic and heating reduction measures on urban air quality. *Atmos. Environ. 29,* 3477-3487.

*Bartzokas A. and Metaxas D.A.*, 1993: Covariability and climatic changes of the lower troposphere temperatures over the Northern Hemisphere. *Il Nouvo Cimen 16C,* 359-373.

*Chin, A.T.H.*, 1996: Containing air pollution and traffic congestion: transport policy and the environment in Singapore. *Atmos. Environ., 30,* 787-801.

*Embrechts, P.*, 1997: *Modelling Extremal Events.* Springer-Verlag Berlin, Heidelberg.

*Fang, S.H. and Chen, H.W.*, 1999. Air quality and pollution control in Taiwan. *Atmos. Environ. 30,* 735-741.

*Hastie, D.R., Shepson, P.B., Reid, N., Roussel, P.B. and Melo, O.T.*, 1996: Summertime $NO_x$, $NO_y$ and ozone at a site in rural Ontario. *Atmos. Environ. 30,* 2157-2165.

*Kassomenos, P., Kotroni, V. and Kallos, G.*, 1995: Analysis of climatological and air quality observations from greater Athens area. *Atmos. Environ. 29,* 3671-

*Kassomenos, P., Skouloudis, A.N., Lykoudis, S. and Flocas, H.A.*, 1999: "Air quality indicators" for uniform indexing of atmospheric pollution over large metropolitan areas. *Atmos. Environ. 33,* 1861-1879.

*Makra, L.*, 1999: Concentration of contaminating gases and their connection with large-scale weather situations in Szeged. *The Association of American Geographers, 95th Annual Meeting,* Honolulu, Hawaii, 23-27 March 1999. *Abstracts* 376.

*Makra, L. and Horváth, Sz.*, 1999: Spatial and temporal characteristics of air quality status over a middle-sized urban area. *The 15th International Congress of Biometeorology (ICB'99) and the International Conference of on Urban Climatology (ICUC'99)*, Sydney, Australia. 8-12th November 1999. *Proceedings*. ISBN 1 86408 543 6

*Makra, L., Tar, K. and Horváth, Sz.*, 2000a: Some statistical characteristics of the wind energy over the Great Hungarian Plain. *The International Journal of Ambient Energy 21*, 85-96.

*Makra, L., Horváth, Sz., Zempléni, A., Csiszár, V., Tar, K., Motika, G., Sümeghy, Z. and Károssy, Cs.*, 2000b: Spatial and temporal characteristics of air qualitiy status in southern Hungary. *3rd European Conference on Applied Climatology, ECAC 2000*, Pisa, Italy. 16-20th October, 2000. *Proceedings*. CD Session VI.

*Morawska, L., Thomas, S., Bofinger, N., Wainwright, D. and Neale, D.*, 1998: Comprehensive characterization of aerosols in a subtropical urban atmosphere: particle size distribution and correlation with gaseous pollutants. *Atmos. Environ. 32*, 2467-2478.

*Reiss, R-D. and Thomas, M.*, 1997: *Statistical Analysis of Extreme Values*. Birkhhäuser Verlag, Basel.

*Sillman, S.*, 1999: The relation between ozone, $NO_x$ and hydrocarbons in urban and polluted rural environments. *Atmos. Environ. 33*, 1821-1845.

*Spicer, C.W., Buxton, B.E., Holdren, M.W., Smith, D.L., Kelly, T.J., Rust, S.W., Pate, A.D., Sverdrup, G.M. and Chuang, J.C.*, 1996: Variability of hazardous air pollutants in an urban area. *Atmos. Environ. 30*, 3443-3456.

*Tripathi, B.D., Chaturvedi, S.S. and Tripathi, R.D.*, 1996: Seasonal variation in ambient air concentration of nitrate and sulfate aerosols in a tropical city, Varanasi. *Atmos. Environ. 30*, 2773-2778.

*Unger, J.*, 1997: Some features of the development of an urban heat island. *Studia Universitatis Babes-Bolyai, Geographia 42*, 125-131.

*Weissman, I.*, 1977: On location and scale functions of a class of limiting processes with application to extreme-value theory. *Annales of Probabilities 3*, 178-181.