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# Variations of traffic related air pollution on different time scales in Szeged, Hungary and Freiburg, Germany

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## ABSTRACT

Economic activities and everyday life may create weekly variations in concentrations of air pollutants in urban settings. The present study contributes to this experience on the example of two typical mediumsized towns in Central Europe, Szeged and Freiburg considering the following air pollutants: NO, NO<sub>2</sub>, O<sub>3</sub>, O<sub>x</sub> and PM<sub>10</sub>. Five-year data sets of hourly observations (1997–2001) collected in downtown traffic junctions are analysed. In addition, the effect of the weekly variation on the diurnal course of the air pollutants is also demonstrated, which is especially important when we consider the possible extremes of these traffic related air pollutants. Since the annual variation of the pollutants explains only a minor part of the total variance and, furthermore, the weekly variation behaves rather similarly in the different seasons, the weekly variation of the diurnal peaks is quantified for the whole year. The average annual variations of NO, NO<sub>2</sub>, O<sub>3</sub> and O<sub>x</sub> are very similar for both Szeged and Freiburg. Annual levels of NO<sub>2</sub> and O<sub>3</sub> are moderately higher, while those of  $PM_{10}$  are extremely higher in Szeged, which is reflected in their average weekly and diurnal variations, too. In Freiburg the diurnal variation of PM10 shows a clear daily course with only one wave, compared to that for Szeged with the shape of a double wave. In Szeged, highest percentile values of NO and NO<sub>2</sub> occur mostly in the evening, while in Freiburg either in the mourning or in the evening and generally there is very little difference between them. In Szeged, maximum of  $O_3$ peak values, while in Freiburg minimum of them are found on weekends.

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# 1. Introduction

Air pollution is one of the most important environmental problems, which is restricted mostly to the cities. Generally, human activities induce monotonous accumulation of pollutants. Population growth in cities and, in connection with this, the increase of built-up areas is considered to be some of the underlying reasons for worsening air quality. A considerable part of population growth derives from migration to the cities. The ever-increasing urban population, together with the growing industrialisation and energy consumption, and the extensive transportation, increase air pollution, which becomes a more and more serious challenge for the interest of survival. The main sources of air pollution are industrial activity, motor vehicle traffic (which heavily affects air quality in densely urbanised regions) and emissions from building heating systems (contribution of which is important in the winter period).

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*E-mail addresses*: makra@geo.u-szeged.hu (L. Makra), helmut.mayer@meteo. uni-freiburg.de (H. Mayer), mika.j@met.hu (J. Mika), santa.tamas@mailbox.hu (T. Sánta), jutta.holst@meteo.uni-freiburg.de (J. Holst). 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 the size of the city, as well as various physical and chemical processes (industrial activity), meteorological processes (climate, local meteorological conditions at the moment), geographical processes (structure and quality of the surface, vegetation cover, position, relief) and social factors (existing environmental regulations, urban planning policies) (Mayer, 1999).

Air pollutants can be divided into two groups: the traditional Major Air Pollutants (MAPs) and the Hazardous Air Pollutants (HAPs). The MAPs include sulphur dioxide, nitrogen dioxide, carbon monoxide, particles, lead and the secondary pollutant ozone, while the HAPs include chemical agents [e.g. volatile organic compounds (VOC), benzene, polyaromatic hydrocarbons (PAH)], physical agents (e.g. depositing dust coming from the surface, yellow dust in China forming loess) and biological agents (e.g. pollen of plants). The HAPs are generally present in the atmosphere in much smaller concentration than the MAPs and they often appear more localised, but they are – due to their high specific activity – nevertheless toxic or hazardous. Both in scientific investigations and in

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abatement strategies HAPs are difficult to manage not only because of their low concentrations, but also because they are in many cases not identified (Fenger, 1999).

Research on urban air has a wide literature. Some of these are concerned with the analysis of characteristics of pollutants (e.g. Ol-cese and Toselli, 2002), others deal with the spatial and temporal variability of those (e.g. Mayer and Haustein, 1994; Hunová et al., 2004; Rost et al., 1999), or investigate statistical interrelationships among the variability of pollutants (e.g. Qin et al., 2004), or examine social policy on regulating emissions (e.g. Fehrenbach et al., 2001), or study the connection of meteorological parameters with chemical/biological air pollutants (e.g. Raga et al., 2001/Makra et al., 2004), or evaluate urban air quality using special models (e.g. Jorquera, 2002a, 2002b) and special air quality indicators (e.g. Mayer et al., 2004).

Motor vehicle traffic seems to be one of the most important sources of air pollution, mainly in the cities. Cities in Hungary, especially the big ones like Szeged, together with the population, and the number of motor vehicles, are continuously expanding. Consequently, a more significant role should be assigned for traffic as potential air quality influencing factor in the future (Ministry for Environment, Hungary, 1999).

In Hungary, motor vehicle traffic related emissions of CO, NO<sub>x</sub> and PM<sub>10</sub> are around 70%, 55% and 14%, respectively, of the total emissions of these pollutants (Ministry for Environment, Hungary, 1999).

The traffic system of Szeged is considered to be overcrowded. Among vehicles, taking part in the traffic, the ratio of passenger cars is the highest (84%). During the 11-year period between 1990 and 2000, traffic density has not changed considerably. However, the types of vehicles have been fundamentally modified. Emission from more and more vehicle types decreases significantly, as the ratio of vehicles equipped with catalytic converter keeps increasing. Hence, in contrast to stagnant traffic density, emissions of vehicle related air pollutants have been decreasing considerably. This way, e.g. CO emissions of motor vehicles in the year 2000 were only 35–40% of those measured in 1990 (Pitrik, 2000).

The objectives of the present study are: to determine the typical diurnal, weekly and annual variations of the traffic related air pollutants NO, NO<sub>2</sub>, O<sub>3</sub>, O<sub>x</sub> and PM<sub>10</sub> in Szeged and Freiburg, to determine statistical interrelationships of these air pollutants in each city, to compare the temporal course of the pollutants between the cities, furthermore, as a case study, to analyse their relation to meteorological elements for Szeged, by using the dataset of a self-performed traffic census within the 5-year period analysed (1997–2001).

Average diurnal, weekly and annual cycles of pollutant gases and their percentile values have already been analysed in the literature (Mayer, 1999). However, apart from this analysis, the study represents the above characteristics for  $PM_{10}$ ; furthermore, interrelationship of traffic density, pollutant levels and meteorological elements and, besides, it makes a comparison of the temporal course of the pollutants between Szeged and Freiburg. Our aim to choose Freiburg was, on the one hand, that both cities are found in Central Europe (Szeged in the eastern part and Freiburg in the western part of Central Europe), on the other, that both are considered middle-sized cities with nearly the same size and population, however, with definitely different development level of the infrastructure.

#### 2. Data and methods

## 2.1. Location, structure and climate of Szeged and Freiburg

Szeged ( $\varphi = 20^{\circ}06'$ E;  $\lambda = 46^{\circ}15'$ N; h = 79 m a.s.l.) lies near the confluence of the Tisza and Maros Rivers. It is one of the largest cit-

ies in Hungary with 155,000 inhabitants covering an area of about 46 km<sup>2</sup> (Fig. 1). Szeged and its surroundings occupy a flat and open region and the city has the lowest elevation value in Hungary.

The total urban spread extends well beyond the city limits and includes the largest oil field in Hungary with several oil torches, just north of the town. This oil field is a significant source of  $NO_x$  and sulphur dioxide. The power station, located in the north-western part of the town, and motor vehicle emissions have largely contributed to the nitrogen oxide levels at Szeged.

The mean annual temperature is 11.2 °C, while the mean January and July temperatures are -1.2 °C and 22.4 °C, respectively. As for yearly averages, annual precipitation is 573 mm, relative humidity is 71%, wind speed is 3.2 m s<sup>-1</sup>, and sunshine duration is 2102 h (1961–1990).

The basis of the city structure is a boulevard-avenue street system cut through by the Tisza River. This way, though the structure of the city is simple; however, due to this system, motor vehicle traffic as well as air pollution is concentrated to the downtown areas. The industrial area is located mainly in the north-western part of the town. Thus, the prevailing westerlies and northerlies transport pollutants, originating from this area, towards the centre of the city.

Freiburg ( $\varphi$  = 7°50′E;  $\lambda$  = 48°00′N; h = 240 m a.s.l.) is a mediumsized city, without heavy industrial sites (Fig. 1). Freiburg is located in the south-western corner of Germany near the French and Swiss borders. The city, located at the eastern border of the southern upper Rhine valley directly at the foot of the low mountain range "Black Forest", covers an area of some 150 km<sup>2</sup>. At present, Freiburg has a growing population of approximately 200,000 people.

The mean annual air temperature is  $10.7 \degree C$  (1961-1990), while maximum and minimum monthly mean temperature are  $19.9 \degree C$  in July (1961-1990) and  $1.8 \degree C$  in January, respectively. The annual precipitation total amounts to 956 mm (1991-1990). The mean annual relative humidity is 73%, mean sunshine duration 1808 h. Due to its geographical location within the Rhine valley, wind speeds are generally low (the annual mean wind speed is  $2 \text{ m s}^{-1}$ ). During clear nights a regional wind system ("Höllentäler") forms with phases of enhanced wind speeds from the east.

## 2.2. Measurement of the pollutants considered

The database consists of 30-min averages of the air pollutants examined. Average daily mass concentrations of NO, NO<sub>2</sub>, O<sub>3</sub> and PM<sub>10</sub> ( $\mu$ g m<sup>-3</sup>) are used for the 5-year period between 1997 and 2001. The data come from the monitoring stations in Szeged and Freiburg downtown, located at traffic junctions (Fig. 1).

#### 2.3. Traffic census in Szeged

Peak hours at the traffic junction, Szeged (Kossuth Lajos boulevard – Damjanich street and Teréz street) are at about 8–10 a.m., 1–2 p.m. and 4–5 p.m. On a weekday, about 30,000 (84%) motor vehicles and vans, 1500 (4%) buses, 3000–3500 (8.4–9.8%) lorries, 900 (3%) motor bikes and 20 (0.06%) low speed vehicles (e.g. tractor) proceed through the crossing here. At the time of the census a great number of motor cars (38%, as an average for Hungary) were equipped with exhaust catalysers.

In order to clarify potential effect of traffic to the concentration of air quality parameters, measured at the monitoring station, traffic census was performed at the crossing during a 1-day period, from 9<sup>30</sup> a.m. September 12, until 9 a.m. September 13, 2000. During this period (calm weather, undisturbed radiation conditions) a continuous traffic census was made and the 30-min totals of motor cars were recorded according to vehicle types. Technical background of the traffic census was adopted from the procedure used by the State Highway Managing Public Utility Company of

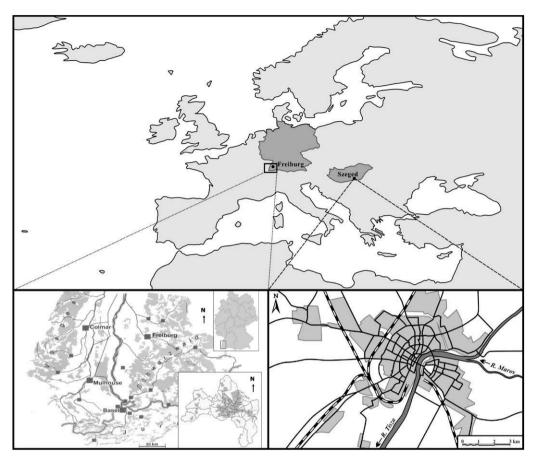


Fig. 1. Geographical position of Szeged, Hungary (source: Department of Climatology and Landscape Ecology, University of Szeged) and Freiburg, Germany (source: Institute for Human Geography, University of Freiburg).

Csongrád County. During the traffic census vehicle types were separated, which (with their so called "vehicle unit factors" in inhabited areas) were as follows: motor vehicle and van (1.0), single public bus (1.8), articulated public bus (2.5), light lorry (1.4), single heavy lorry (1.8), trailer truck (2.5), motor bike (0.7) and slow motor vehicles (e.g. tractor) (2.5). On the basis of the traffic census, the number of motor cars within each vehicle type was multiplied with their own factors and then the results were summarized for each vehicle type. In this way, vehicle factor totals were received in each 30-min period during this 1-day (24-h) time interval. The time series of this vehicle factor totals was one of our databases. The vehicle unit factor refers to the role of the given vehicle in traffic; namely, it shows that what kind of role the given vehicle plays in decelerating or accelerating traffic, relative to a motor vehicle or van. In this way, the vehicle factor totals do not relate directly to the concentration of the air guality parameters; however, this factor might be in significant correlation with that.

# 3. Results

# 3.1. Comparison of the annual variation of the pollutant concentrations and its effect on variability

For Szeged, the diurnal concentrations of NO display a clear annual variation, while those of  $NO_2$  and  $PM_{10}$  indicate less definite ones with much higher fluctuation around this variation. However, annual variations for all three of them are characterised by winter maxima and summer minima. In contrast, the diurnal concentrations of  $O_3$  with a clear annual variation have a winter minimum and a summer maximum (Fig. 2).

The average weekly variations of the air pollutants for Szeged, after the annual variation had been removed, were also determined. For this, three seasons were defined: Winter (WI) (November–February); Summer (SU) (May–August) and a Transient Period (TR) (March–April and September–October). The average weekly variations of NO, NO<sub>2</sub> and PM<sub>10</sub> are very similar with weekday maxima and weekend minima. The highest concentrations are observed in the winter, while the lowest ones in summer. On the other hand, that of O<sub>3</sub> has weekday minima and weekend maxima. On weekdays the concentration of traffic related NO is high. After reacting with O<sub>3</sub>, its concentration is decreased:

$$\mathrm{NO} + \mathrm{O}_3 \to \mathrm{NO}_2 + \mathrm{O}_2 \tag{1}$$

Conversely, at the weekend, the concentration of  $O_3$  is high, due to the relatively low traffic.

Fig. 3 compares the average annual variations of NO, NO<sub>2</sub>, O<sub>3</sub>, O<sub>x</sub> and PM<sub>10</sub>. O<sub>x</sub> is a measure of the O<sub>3</sub> concentration, contained in an air mass. It is defined as the sum of NO<sub>2</sub> and O<sub>3</sub> and is more suitable for the assessment of the photochemical O<sub>3</sub> budget than O<sub>3</sub> alone, because it takes the reversible chemical processes into account as well (Mayer, 1999). Due to the topographically caused, reduced atmospheric exchange conditions in the southern upper Rhine valley in winter, the annual variation of the primary air pollutant NO displays higher extremes for Freiburg (higher values in the winter months and lower in the rest part of the year) than that for Szeged (Fig. 3). As the NO concentration depends not only on the rate of emission, but on the prevailing weather conditions as well, higher winter values refer to atmospheric stability with frequent inversions. The average annual variation of NO has the lowest values during the summer (June and July), when air mass

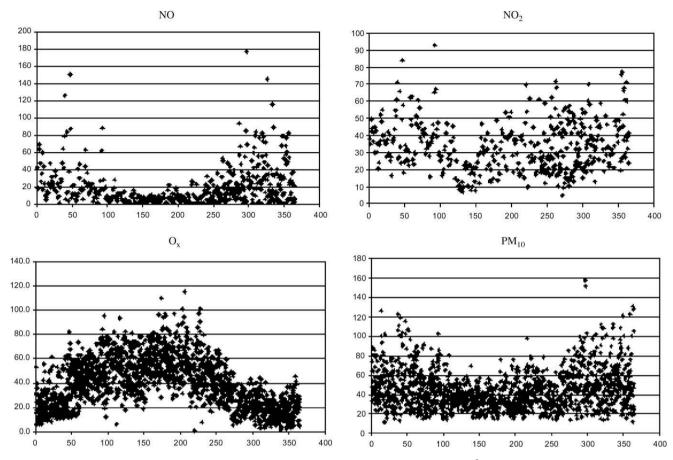


Fig. 2. Annual variations of diurnal mean concentration for NO, NO<sub>2</sub>, O<sub>3</sub> and PM<sub>10</sub> (µg m<sup>-3</sup>), Szeged, 1997–2001.

exchange is the most intensive. The annual variation of NO<sub>2</sub>, a secondary substance produced mainly by chemical reactions, follows a similar course to that of NO and shows higher values for Szeged than for Freiburg (Fig. 3). Tropospheric ozone is produced via the effect of short-wave radiation on substances emitted from anthropogenic sources. The role of solar radiation in the troposphere, producing photochemical O<sub>3</sub> can be expressed by the following pair of chemical equations:

$$\begin{array}{l} \mathsf{NO}_2 + h\nu \to \mathsf{NO} + \mathsf{O} \\ \mathsf{O} + \mathsf{O}_2 + \mathsf{M} \to \mathsf{O}_3 + \mathsf{M} \end{array} \tag{2}$$

(h: Planck-constant; v: frequency of irradiance; M: usually a molecule of O<sub>2</sub> or N<sub>2</sub>) (Sindosi et al., 2003). [Simple chemical reactions of (1) and (2) influence daily variations of NO, NO<sub>2</sub> and O<sub>3</sub>.] Consequently, the average annual variation of O<sub>3</sub>, together with that of O<sub>x</sub>, has the greatest values in the summer (June and July) (Makra et al., 2001) (Fig. 3). In Szeged the average annual variation of PM<sub>10</sub> has the greatest values in November, December and January, while the lowest ones in the summer months (Fig. 3). Higher winter values might refer to atmospheric stability with frequent inversions. The lowest values during the summer (June, July, August and September) can be explained by dilution caused by intensive vertical exchange in the atmosphere. On the other hand, in Freiburg average annual variation of PM<sub>10</sub> is much less pronounced and well below the values of Szeged. This is mainly due to the fact that the annual radiation is lower (by 300 h) and the annual precipitation is higher (by 400 mm) (see Chapter 2), hence aridity index is well below one in Freiburg, while it exceeds considerably the value one in Szeged. In this way there is less dust in Freiburg to get into the air either by winds or by traffic (Fig. 3).

# 3.2. Comparison of the weekly and diurnal variations of the pollutant concentrations

In both cities the diurnal variations of NO and NO<sub>2</sub> have the shape of a double wave, with bigger amplitudes for NO than for  $NO_2$  (Fig. 4). Due to the traffic density, the concentration of NO is relatively higher on weekdays, than on weekends. This effect can also be observed for the secondary substance NO<sub>2</sub>. The average diurnal variations on weekdays are greater for NO than for NO<sub>2</sub>, because NO<sub>2</sub> has a longer lifespan than the more reactive NO. Generally, the NO concentrations are higher in the morning, then in the evening. This can be explained by the fact that in the morning the rush hour is shorter, and the atmosphere near the surface is more stable than in the evening. The low NO concentrations early in the afternoon result mainly from the reduction of O<sub>3</sub> by NO. In Freiburg, NO values show higher first maxima and lower second maxima and second minima on weekdays, while lower ones on weekend compared to those in Szeged (Fig. 4). NO<sub>2</sub> concentrations in Freiburg are well below than those in Szeged. Moreover, in Freiburg the first maximum of NO<sub>2</sub> values is in the mourning rush hours, while the second maximum in the evening, just opposite to those in Szeged (Fig. 4). The diurnal variations of O<sub>3</sub> show a clear daily course with one wave in both cities but with higher values for Szeged (Fig. 4). A maximum takes place in the early afternoon caused by photochemical O<sub>3</sub> formation, while a minimum occurs after midnight. On the basis of its definition, the diurnal variation of  $O_x$  is similar to that of  $O_3$ . Both in Szeged and Freiburg on

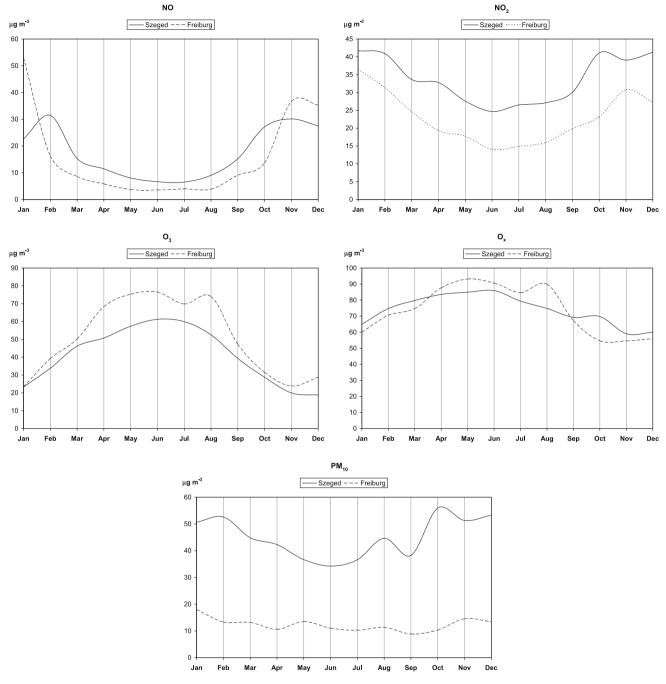


Fig. 3. Comparison of average annual variations of NO, NO<sub>2</sub>, O<sub>3</sub>, O<sub>x</sub> and PM<sub>10</sub> monitoring stations, Szeged and Freiburg, 1997–2001 (NO, Freiburg, 1997–1998).

weekends the average  $O_3$  maximum values are a little higher than on weekdays, but this is not valid for  $O_x$  (Makra et al., 2001). The diurnal variations of  $O_x$  are just the same in the two cities (Fig. 4).

In Szeged the weekly and diurnal variations of  $PM_{10}$  have the shapes of double waves (Fig. 4). Both primary and secondary maxima can be observed during peak hours and, in the same way, primary and secondary minima occur, when traffic is the lowest (at night) or is decreasing (around midday). Also, due to the dense traffic, the concentration of  $PM_{10}$  is relatively higher on weekdays and lower on weekend. However, in Freiburg the concerning curve of  $PM_{10}$  is quite different showing a clear daily course with only one wave (Fig. 4). It is evident that in Freiburg the weekly and diurnal variations of  $PM_{10}$  are not traffic dependent.

Analysing NO and NO<sub>2</sub> peak values, it becomes obvious that in Szeged the highest values occur most frequently in the evening (Fig. 5), while the diurnal variation of mean NO concentrations has its local maximum in the morning (Fig. 5). On the other hand in Freiburg the highest values occur either in the mourning or in the evening and generally there is very little difference between them (Fig. 5). In Szeged, maximum of O<sub>3</sub> peak values are found on weekends, while in Freiburg there is a slight increase in the mean values, and the peak values do not differ as significantly as on weekdays (Fig. 5).

Peak values for  $O_3$  and  $O_x$  show clear daily courses with one wave in both cities but with higher values for Szeged, similarly to their curves of weekly and diurnal variations (Fig. 5).

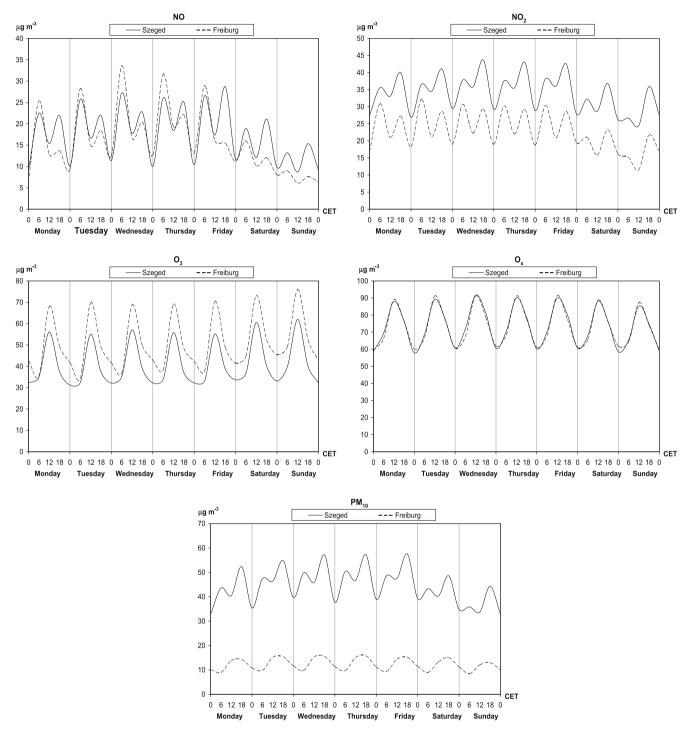


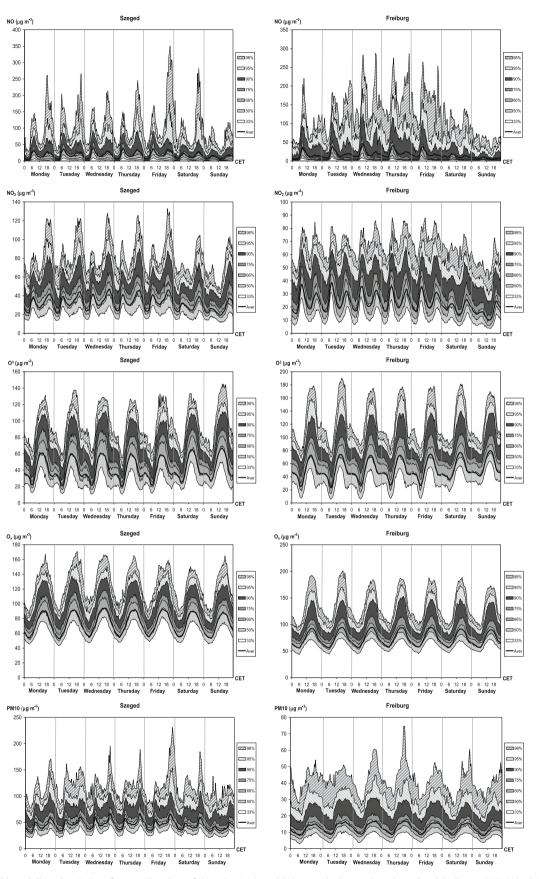
Fig. 4. Comparison of average weekly and diurnal variations of NO, NO<sub>2</sub>, O<sub>3</sub>, O<sub>x</sub> and PM<sub>10</sub> monitoring stations, Szeged and Freiburg, 1997–2001 (NO, Freiburg, 1997–1998).

In Szeged, peak values for  $PM_{10}$  show a double wave displaying maxima late in the evening, while secondary maxima can be observed late in the morning. The lowest  $PM_{10}$  concentrations are measured early in the morning, while secondary minima occur in the evening (Fig. 5). The average annual, weekly and diurnal variations, as well as the average weekly and diurnal variations of the percentile values of  $PM_{10}$  are very similar to those of NO, which indicates the relation of  $PM_{10}$  to traffic densities. In Freiburg, peak values for  $PM_{10}$ , contrary to those for Szeged, indicate one wave daily, generally with evening maxima (Fig. 5).

#### 3.3. Traffic density, pollutant levels and meteorological elements

Vehicle factor totals were related to pollutant levels and meteorological elements for the 1-day traffic census ( $9^{30}$  a.m. September 12–9 a.m. September 13, 2000) in Szeged.

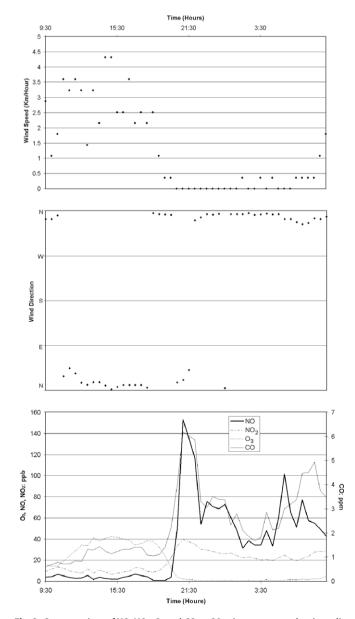
Analysis of 30-min average concentrations of  $O_3$  shows its values between 25 and 70 µg m<sup>-3</sup> (Fig. 6).  $O_3$  peaks are well below the information threshold of the EC Directives (1 h  $O_3$  threshold value is 180 µg m<sup>-3</sup>, which corresponds to 86.6 ppb (basis of the conversion: t = 20 °C, p = 1013 m b; Fig. 6). Maximum  $O_3$  concentrations occurred around noon. At the same time, in the only period (be-



**Fig. 5.** Average weekly and diurnal variation of percentile values of NO, NO<sub>2</sub>, O<sub>3</sub>, O<sub>x</sub> and PM<sub>10</sub>, monitoring stations, Szeged and Freiburg, 1997–2001 (NO, Freiburg, 1997–1998).

tween 9<sup>30</sup> a.m. and 8<sup>00</sup> p.m.) when breeze was observed (north and north-east winds), NO<sub>2</sub>/NO ratio was greater then unity. Whereas, in the evening and at night the situation is reverse. In this latter case NO<sub>2</sub>/NO < 1. However, ratio of NO<sub>2</sub>/NO depends not primarily on wind speed but, through ozone concentration, on radiation and NO emissions. Daytime, the ratio NO<sub>2</sub>/NO > 1 can be explained by the rapid oxidation of NO (NO + O<sub>3</sub>  $\rightarrow$  NO<sub>2</sub> + O<sub>2</sub>) (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). This is independent of decrease in wind speed resulting from energetic reasons, generally in the evening hours (Bogo et al., 1999; Matzarakis et al., 1999) (Fig. 6).

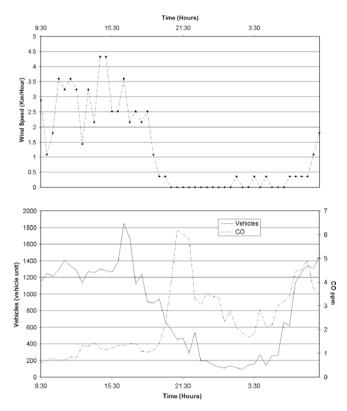
On the day of the traffic census there were moderate northerly winds from  $9^{30}$  a.m. until 8 p.m. Beyond this period no winds blew. Daily courses of CO and NO show characteristic synchronous changes (Figs. 6 and 7). Their correlation coefficient: r = 0.919, which is significant at the 99.9% probability level (Fig. 8). As CO



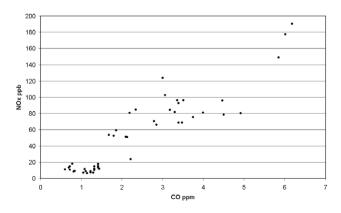
**Fig. 6.** Concentrations of NO, NO<sub>2</sub>, O<sub>3</sub> and CO, as 30-min averages, at the air quality monitoring station, Szeged downtown, from  $9^{30}$  September 12 until  $9^{00}$  September 13, 2000. Wind speed and wind direction are also shown.

and NO are dominantly of traffic origin, daily course of their concentrations and that of the vehicle unit factor might be expected similar (Figs. 6 and 7). However, during daytime they show opposite relation and their courses are parallel only during night and early in the morning (Figs. 6 and 7). The daytime temperate breeze reduces CO and NO levels, at the same time during night, when stability of the atmosphere increases (wind speed decreases), CO and NO concentrations increase in the surface layers, contrary to the decreasing emissions.

The concentration of CO and NO<sub>x</sub> correlates strongly (r = 0.919) during the day of the traffic census (Fig. 8). Values of CO/NO<sub>x</sub> ratio indicate important role of traffic (Makra and Horváth, 1999).



**Fig. 7.** Traffic density and concentration of CO, as 30-min averages at the air quality monitoring station, Szeged downtown, from 9<sup>30</sup> September 12 until 9<sup>00</sup> September 13, 2000. Wind speed is also shown.



**Fig. 8.** Concentration of CO and NO<sub>x</sub>, as 30-min averages at the air quality monitoring station, Szeged downtown, from 9<sup>30</sup> September 12 until 9<sup>00</sup> September 13, 2000.

Pollutant	CO		NO <sub>2</sub>		NO <sub>x</sub>		SO <sub>2</sub>		PM10	
Standard <sup>d</sup> Limit value	<sup>ь</sup> н 3000	°EC	<sup>ь</sup> Н 40	<sup>c</sup> EC 40	<sup>ь</sup> Н 100	<b>°ЕС</b> 30	<sup>ь</sup> Н 50	<sup>c</sup> EC 20	<sup>ь</sup> н 40	<sup>с</sup> ЕС 30
Szeged	° <b>175.0</b>	-	84.7	84.7	51.5	<sup>e</sup> 171.6	9.9	24.8	e <b>112.8</b>	° <b>150.4</b>
Freiburg	-	-	-	57.3	-	°130.3	-	24.6	-	41.3

<sup>a</sup> NO, Freiburg, 1997–1998.

<sup>b</sup> H: limit values, Hungarian Standard (Bozó et al., 2001).

<sup>c</sup> EC: limit values, SO<sub>2</sub>, NO<sub>2</sub>, NO<sub>3</sub>, PM: Council Directive, 1999/30/EC (1999. 04. 22.); CO: Council Directive, 2000/69/EC (2000. 11. 16.), O<sub>3</sub>: Council Directive, 2002/3/EC (2002. 02. 12.).

 $^{d}$  µg m<sup>-3</sup>.

Table 1

Bold: exceeding of the limit values.

For further calculations, mean annual concentrations of the pollutants were computed for Szeged and Freiburg and were expressed in unit of the limit values (Table 1). As a result, in both cities  $SO_2$  levels are the lowest, which meet the standard of the EC Directives. On the other hand, for Szeged concentrations of CO,  $NO_x$  and  $PM_{10}$  exceed the limit values at least 50%, which clearly indicates the role of traffic, while for Freiburg only  $NO_x$  levels exceed the limit.

## 4. Conclusions

The new findings of the study can be summed up as follows:

- In Szeged, the average annual variations of NO, NO<sub>2</sub> and PM<sub>10</sub> (with maxima in winter) are opposite to those of  $O_3$  and  $O_x$ (with maxima in summer). The higher winter values are caused by atmospheric stability with frequent inversions. The lowest values in summer are due to dilution caused by intensive vertical exchange in the atmosphere. The highest intensities of photochemical O<sub>3</sub> formation are observed during the early afternoon and the summer. The annual variation of the primary air pollutant NO displays higher extremes for Freiburg than that for Szeged. Furthermore, in Freiburg average annual variation of PM<sub>10</sub> is more uniform with well lower values to those of Szeged. This is mainly due to the fact that the aridity index is well below one in Freiburg, while it exceeds considerably the value one in Szeged. Both Szeged and Freiburg are free of industries. On the other hand, the amount of cars and especially trucks are lower in Freiburg than in Szeged. In this way, apart from geographical and climate factors, there is less dust in Freiburg to get into the air by traffic.
- After removing the annual variations, the very similar average weekly variations of NO, NO2 and PM10 for Szeged show weekday maxima and weekend minima. Oppositely, those of  $O_3$ show weekday minima and weekend maxima. In Freiburg, NO values show higher extremes on weekdays, while lower ones on weekend compared to those in Szeged. NO<sub>2</sub> concentrations in Freiburg are well below than those in Szeged. The diurnal variations of O3 show a clear daily course with one wave in both cities but with higher values for Szeged. Both in Szeged and Freiburg on weekends the average O3 maximum values are a little higher than on weekdays, but this is not valid for  $O_x$ . The diurnal variations of  $O_x$  are just the same in the two cities. In Freiburg the diurnal variation of PM<sub>10</sub> is quite different showing a clear daily course with only one wave. It is evident that in Freiburg the weekly and diurnal variations of PM<sub>10</sub> are not traffic dependent.
- In Szeged, highest percentile values of NO and NO<sub>2</sub> occur mostly in the evening, while those of O<sub>3</sub> show a maximum during the weekends. On the other hand in Freiburg the highest values occur either in the mourning or in the evening and generally

there is very little difference between them. In Szeged, maximum of  $O_3$  peak values, while in Freiburg minimum of them are found on weekends. The here mentioned difference can be traced back to the fact that, though there are no industrial sources in either of the cities, however traffic density in Szeged is higher than in Freiburg.

- Peak values of PM<sub>10</sub> for Szeged show a double wave (1st maximum: late in the evening; 2nd maximum: late in the morning; 1st minimum: early in the morning; 2nd minimum: in the evening). Temporal variations of the percentile values of PM<sub>10</sub> are very similar to those of NO, which relates PM<sub>10</sub> to traffic. The concentrations of NO and O<sub>3</sub> are also traffic related. On the other hand, in Freiburg, peak values for PM<sub>10</sub>, indicate only one wave daily, generally with evening maxima.
- The above differences in the pollutant characteristics between Szeged and Freiburg can come from the different city structure, traffic structure, difference in environmental standards of the automobiles, difference in age of the automobiles, difference in topography and climate.
- Concentration of both CO and NO are in reverse connection with wind speed. As probably both pollutants are predominantly of traffic origin in the traffic junction examined, their concentrations should be changed synchronously, which meets our expectations.

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