

IDŐJÁRÁS

QUARTERLY JOURNAL
OF THE HUNGARIAN METEOROLOGICAL SERVICE

CONTENTS

<i>Ernő Mészáros:</i>	85
<i>László Haszpra:</i> Measurements of atmospheric carbon dioxide at a low elevation rural site in Central Europe	93
<i>Györgyi Baranka:</i> Near surface ozone concentration evaluation and prediction in Budapest	107
<i>Angéla Anda and Márta Hunkár:</i> The effect of alginite on the production and water use of maize	123
News	137
Contents of journal Atmospheric Environment Vol. 33, Nos. 5-9	139

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New results on the chemical composition of aerosol particles in the atmosphere. Are cloud condensation nuclei produced by the biosphere?

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(Manuscript received 18 February 1999; in final form 7 May 1999)

Abstract—It is generally accepted that cloud condensation nuclei (CCN) consist of the water soluble fine aerosol particles with size below about 1 μm . Former results about the chemical composition of inorganic aerosol particles indicated that water soluble compounds were mostly composed of ammonium sulfate, ammonium bisulfate or sulfuric acid as a function of ammonia available in the air. Under continental conditions nitrate can also be found in the particles in a smaller quantity. On this basis it was postulated that CCN were sulfate particles formed in the air by chemical reactions. Under oceanic conditions the precursor gas is dimethyl sulfide, while over the continents sulfate particles originate from sulfur dioxide. Since this gas is mostly of anthropogenic origin, one concluded that continental CCN are mainly due to human activities. Owing to the spectacular progress in analytical chemistry, during the last years the organic part of the atmospheric aerosol has also been studied. The results obtained are of interest in particular when the water soluble fraction of organic compounds is also investigated. These new research efforts resulted in the following findings. Firstly, under continental conditions the organic carbon concentration is comparable to that of sulfate ions. Secondly, at least half of the mass of organic carbon can be found in compounds soluble in water. Finally, an important part of water soluble organic carbon consists of macromolecular compounds. The aim of this paper is to summarize the results of these recent studies, including those carried out in Hungary. On the other hand, the significance of the new information is discussed from the point of view of water vapor condensation. It is speculated, but not proved, that we cannot exclude the possibility that, beside sulfates, organic substances also provide active cloud condensation nuclei.

1. Introduction

The chemical composition of atmospheric aerosol particles is of interest for many problems in environmental science. For atmospheric research the particle composition is important in particular for understanding the phase transition of

water in the air. Thus, cloud droplets form on aerosol particles called cloud condensation nuclei (CCN). Thermodynamic calculations show that water soluble particles are very active in this process. For this reason many programs have been carried out to study the size and nature of water soluble substances. This is understandable since the number of CCN active at a given supersaturation determines the structure of clouds (the concentration and diameter of cloud droplets). This means that they control, among other things, the precipitation forming ability and albedo of the clouds.

Studies done in this field indicated that the number of coarse particles (with diameter above $1\ \mu\text{m}$), formed by the desintegration of the Earth surface, is too low compared to the concentration of cloud droplets. This implies that the sizes of CCN are in the fine particle size range consisting of particles with diameter below the above value. *Junge* (1963) was the first who demonstrated that in continental environment water soluble particles in the size range of $0.2 < d < 2.0\ \mu\text{m}$ (where d is the particle diameter) are composed of ammonium sulfate, ammonium bisulfate or sulfuric acid as a function of gaseous ammonia available. Later it was found (*Mészáros*, 1968) that particles smaller than $0.2\ \mu\text{m}$ contain about half of the mass of sulfate and ammonium ions. This means that, on a number basis, the great majority of sulfate particles have a diameter below the above value. Further, the study of the morphology of fine aerosol particles collected under oceanic conditions made it evident that sulfate particles are predominant in the fine size range even in air above the sea surface (*A. Mészáros and Vissy*, 1974; *Butor*, 1976). These sulfate particles form by the condensation of the reaction products of dimethyl sulfide emitted by the oceanic biota (*Charlson et al.*, 1987). On the other hand, *Twomey* (1968) pointed out on the basis of the volatility of CCN that they consists of sulfate particles. This conclusion was confirmed by several authors using different measuring techniques. Consequently, it was concluded that continental clouds form on sulfate particles of anthropogenic origin (*Twomey*, 1991; *Hudson*, 1991). This raised the question: how did continental clouds form before the industrial revolution (*Mészáros*, 1992). Anyway, it seemed that something is still needed to explain the formation of clouds at least under continental conditions.

2. New findings on the chemical composition

One possible explanation of the formation and structure of continental clouds without human activities is to suppose that CCN are also due to particle production by vegetation. This possibility was raised over three decades ago by *Went* (1960). He postulated that the blue-haze formed over forested areas in summer, during sunny, calm periods, is the result of the reactions of volatile organic compounds released by vegetation. Unfortunately this promising proposition has not been considered in detail after the publication of that paper.

This situation was partly due to analytical difficulties. The second problem was the fact that, before analyses, aerosol samples were generally dissolved in organic solvents. As an example, Rogge *et al.* (1993) leached the samples by hexane and a mixture of benzene/isopropanol and analyzed the liquids obtained by up-to-date gas chromatographic and other techniques. They found that in polluted air in southern California (U.S.A.) 45% of total fine aerosol mass is due to carbonaceous particles. One third of this quantity is elemental (black) carbon, while two thirds is organic carbon. *The ratio of organic carbon to sulfate (measured parallel) is larger than unity.* In spite of the fact that the authors cited identified more than 80 organic compounds, the mass of the species identified constituted only 10% of the total carbon mass. The compounds identified were mostly *organic acids* like aromatic polycarboxylic acids, aliphatic dicarboxylic acids and n-alkanoic acids.

On the basis of their samplings at remote Californian and oceanic sites, Simoneit and Mazurek (1982) stated that higher plant waxes constituted the predominant natural component of the aerosol. These waxes consist mostly of *lipids* with much higher concentrations at continental sites. The materials extractable from lipids by the solvent dichloromethane are composed primarily of n-alkanes (and other hydrocarbons), fatty acids and fatty alcohols. In the air, *carboxylic acids* (simpler than fatty acids) are also ubiquitous. These species have low molecular weights and high polarity. Consequently they are soluble in atmospheric waters. Thus, formic and acetic acids were detected in precipitation samples collected under different conditions. Their concentrations were substantial particularly in the tropics (Keen *et al.*, 1983). Further studies also indicated that carboxylic acids can also be identified in aerosol samples. These studies show that oxalic acid is the most important carboxylic acid in the atmospheric aerosol. The size distribution of oxalate ions is very similar to that of sulfate, which makes it possible that they form internally mixed particles with sulfate ions (Mészáros *et al.*, 1997).

In the atmosphere the natural solvent is water, and lead to phenomena like haze, fog and clouds. For this reason, a new era in the study of organic substances was begun when water soluble substances were also extracted from aerosol samples. Mukai and Ambe (1986) did not use water, but leached the samples by an alkaline solution. By analyzing aerosol samples collected in rural Japan, they obtained in this way a brownish liquid with characteristics very similar to those of *humic acids* common in the soil. This finding was in agreement with the results of Likens *et al.* (1983) indicating that in precipitation samples dissolved macromolecules (molecular mass above 1000) can be found in relatively high concentration. However, Mukai and Ambe reported low concentrations of macromolecular humic-like substances in the atmospheric aerosol.

In Europe, macromolecules in aerosol samples were first identified by Havers *et al.* (1998) in Germany. According to their publication humic-like

substances made up a significant part of organic carbon. These substances consist mainly of polysaccharides and aliphatic substructures. This finding was essentially confirmed by Zappoli *et al.* (1999) on the basis of the analyses of fine aerosol samples collected in Sweden, Hungary and Italy. This international group also demonstrated that an *important part* (half on an average) *of humic-like macromolecules is soluble in water*. The comparison of the thermograms of aerosol particles collected in Hungary and humic acid standards also showed (Gelencsér *et al.*, 1999) that humic-like substances cannot be found in the coarse size range. This means that the formation of the particles containing these materials is different from the simple disintegration of the surface. Recent analyses by pyrolysis gas chromatography-mass spectrometry (Gelencsér, unpublished material) also indicate that macromolecules in the fine size range are composed of polysaccharides, lignins, proteins and lipids very similarly to major soil compounds.

The carbon thermograms of fine aerosol particles collected in rural air over Hungary are characterized by two peaks (Gelencsér *et al.*, 1999). The first peak at lower temperature refers to more volatile and/or more easily oxidizable organic compounds, while the considerably larger second peak represents less volatile and/or less oxidizable species (and a lesser extent elemental carbon). Humic acid standards produced only the second peak at the higher temperature. This indicates that humic acid-like, macromolecular substances constitute the second peak. On the other hand one can speculate that the first peak is mostly due to low molecular mass carboxylic acids. By accepting this concept, which needs further confirmation, one can state that in Hungary the concentration of macromolecular species is much higher than that of carboxylic acids. However, identical analyses of fine aerosol particles sampled in the Swiss Alps (Krivácsy, unpublished material) indicate that in cleaner tropospheric air carboxylic acids account for a higher fraction of organic carbon compared to macromolecular species. This is in agreement with the results obtained recently in very clean Arctic air, according to which water soluble organic species are composed mainly of carboxylic acids (Kawamura *et al.*, 1998).

3. Discussion: the possible composition of CCN

On the basis of the above results one can conclude that an important fraction of fine organic particles is soluble in water. *The water soluble organic species consist either of carboxylic acids with low carbon number or of macromolecular substances that behave like humic acids*. Although carboxylic acids can be formed by chemical reactions in the atmosphere, it is improbable that macromolecular compounds are created in this way. The soil does not produce macromolecular aerosol compounds by mechanical disintegration. This is proved by the fact that no macromolecules were identified in the coarse size

range. Secondly, biomass burning is not substantial in Hungary. For this reason one has to postulate that macromolecular organics in the aerosol are attributed to primary biogenic sources.

It is to be noted that the possibility that natural vegetation emits CCN under tropical and intertropical conditions was proposed several years ago. *Désalmand et al.* (1982) found in the Ivory Coast that in the dry season CCN are due to biomass burning, while in the rainy season high relative humidities, favorable for the bacterial decomposition of plants, result in higher CCN concentrations. Désalmand and her coworkers did not measure the particle composition. However, other studies on the elemental composition carried out in the Amazon basin show that primary biogenic particles constitute 55–95% of the mass of fine particles (*Artaxo and Hansson, 1995*). On the basis of the material presented in this paper we can raise the question whether *organic particles give an important part of CCN even at mid-latitudes*. Considering the large water soluble organic fraction of the atmospheric aerosol we cannot exclude that the answer is positive. Thus, we have to take into account seriously this possibility.

It would be difficult to compare the importance of organic water soluble substances in condensation relative to that of sulfate particles. For example, in rural air over Hungary about one third of the mass of fine particles is composed of sulfate. Organic carbon also constitutes about 30% of the total mass concentration. However, this fraction is expressed as *elemental* carbon and not an *organic compound*. As we have seen above, macromolecular species are present in a rather high proportion. For this reason we can suppose with caution that carbon concentration must be multiplied by two, as a first approximation, to obtain the concentration of carbon compounds as a whole. This speculation results in organic mass two times greater than that of sulfate ions. Since only half of the organics is soluble in water (see above), one receives in this way that *the mass concentration* of water soluble organic species is approximately equal to that of sulfate. One has to note in this respect that *Saxena et al.* (1995) measured the hygroscopic growth of particles in Arizona (U.S.A.) by using a special measuring technique. They also determined the size distribution of ammonium sulfate and organic particles. At the same time they calculated, by a thermodynamic approach, the hygroscopic particle growth due to ammonium sulfate alone. They found that at high relative humidities the water quantity measured was twice the calculated value. Since the organic mass fraction correlated well with the excess water, the American authors concluded that the half of the liquid water on aerosol particles is caused by the presence of water soluble organic compounds. This conclusion is in surprisingly good agreement with the results of our chemical measurements.

One has to also mention that recent fog studies in Po Valley, Italy, demonstrate (*Facchini et al., 1999*) that the partitioning of organic compounds between fog water and interstitial aerosol depends on the water solubility of particles. Thus, *polar water soluble species* constituting the major fraction of

total carbon *can be found mostly in droplet phase*, while insoluble carbon is detected preferentially in interstitial particles. This study also shows water soluble organic species are scavenged by fog droplets similarly to inorganic ions. This important finding also points in the direction that CCN consist at least partly of water soluble organics.

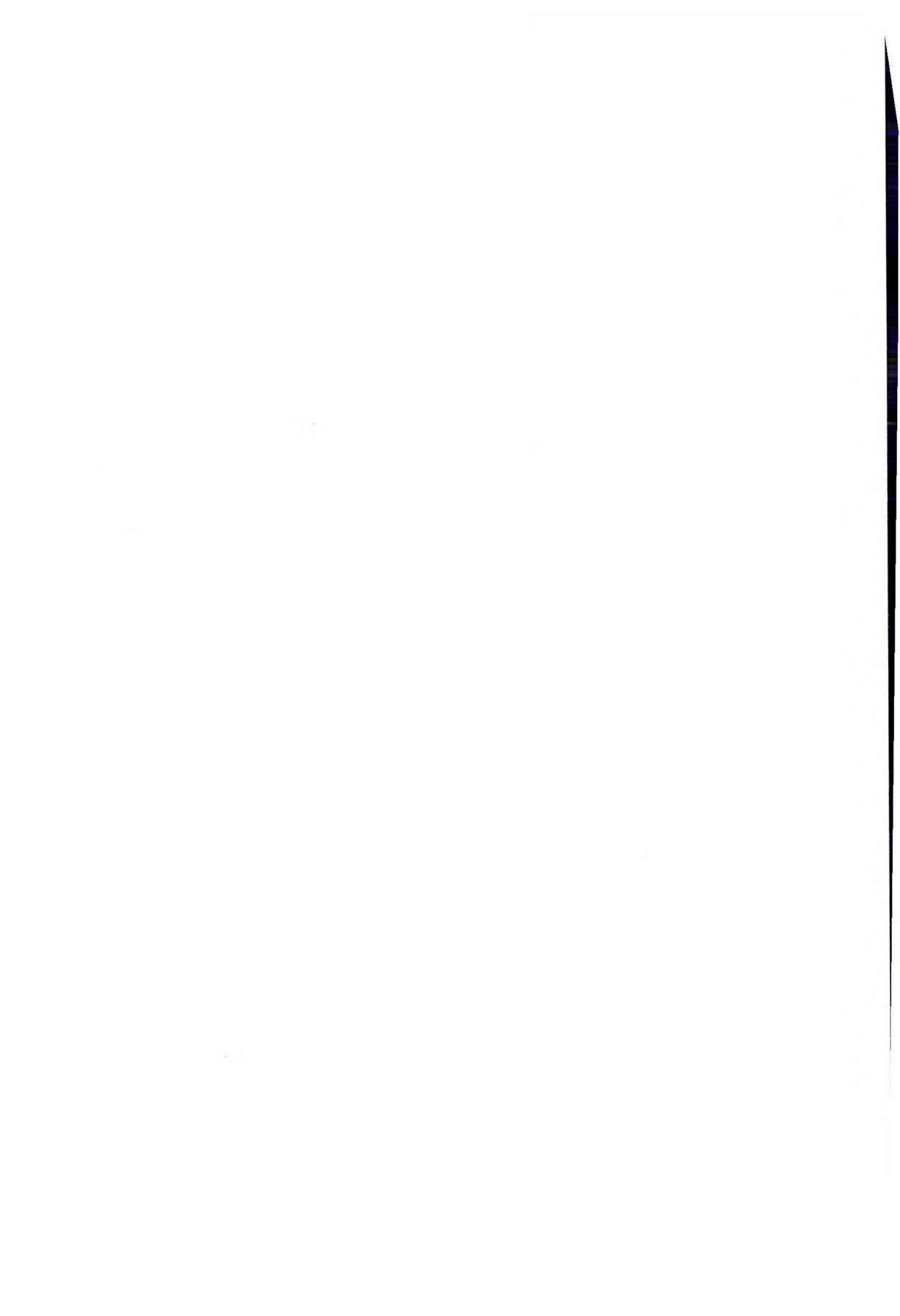
For further considerations it would be necessary to have some information on the size distribution of macromolecular compounds, since size plays an important part in the control of the condensation activity of aerosol particles. Another problem is whether CCN have a sufficient size (mass) to be detected by chemical analyses of fine particles. Anyway we note that there is some indication that organic species constitute smaller particles than sulfate ions (Novakov and Penner, 1993). This means that the *number concentration* of organic particles is higher than the number of sulfate particles if their mass concentrations are equal. This would point in the direction that organic particles give more CCN than sulfate aerosols. However, the solubility of the humic acid-like species is an open question. Although this is not proved, one can speculate that the affinity of sulfate to water is stronger than that of soluble organic species. This means that sulfate particles are active in condensation at lower supersaturations than organic particles.

Finally, we can state that during the last years much new information has been obtained concerning the chemical composition of aerosol particles in the air. This information makes it possible to consider the problem of CCN in a new perspective. However, many new questions have arisen. Thus, it goes without saying, that before drawing a final conclusion on the nature of CCN, further studies are needed. It seems to be particularly important to link direct measurements of CCN with chemical analysis of the organic fraction of the atmospheric aerosol.

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Measurements of atmospheric carbon dioxide at a low elevation rural site in Central Europe

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(Manuscript received 4 January 1999; in final form 20 April 1999)

Abstract—The atmospheric mixing ratio of carbon dioxide has been measured since 1981 at K-puszta (Hungary) as part of the WMO BAPMoN/GAW program. The data show remarkable daily and seasonal variations due to the activity of the surrounding vegetation. The mean daily amplitude is 43 ppm in July and 5 ppm in January. Considering only the spatially more representative daytime values the annual amplitude of the mixing ratio is 28 ppm. The trend and the temporal course of the growth rate of the daytime mixing ratio are similar to those observed at Mauna Loa, Hawaii, Point Barrow, Alaska, or at monitoring sites in Germany. The overall trend during 1981–1998 is 1.51 ppm/year with a short term fluctuation between -4.3 and $+7.0$ ppm/year.

Key-words: trends in the composition of the atmosphere, carbon dioxide, diurnal variation, seasonal variation.

1. Introduction

The atmospheric mixing ratio of carbon dioxide has been increasing for at least 200 years due to human activity (see ice core measurements of e.g. *Barnola et al.* (1995) or *Etheridge et al.* (1996), and the direct atmospheric measurements of e.g. *Beardsmore and Pearman* (1987), *Keeling et al.* (1989), *Keeling and Whorf* (1994), *Thoning et al.* (1994) or *Levin et al.* (1995)). This increase may enhance the greenhouse effect of the atmosphere generating global climatic change (*IPCC*, 1996). Recently, several papers based on atmospheric measurements and model calculations (e.g. *Keeling et al.*, 1989; *Tans et al.*, 1989; 1990; *Enting and Mansbridge*, 1991; *Musselman and Fox*, 1991; *Tans*, 1991; *Quay et al.*, 1992; *Sarmiento and Sundquist*, 1992; *Sundquist*, 1993; *Dixon et al.*, 1994; *Hesshaimer et al.*, 1994; *Ciais et al.*, 1995; *Denning et al.*, 1995; *Taguchi*, 1996) concluded that there should be a large CO₂ sink in the northern hemisphere. A number of these

papers suggest that this “missing sink” is the terrestrial biosphere in the northern temperate latitudes. The characterization and quantification of the “missing sink” needs additional monitoring sites in this region (Tans, 1991) and the evaluation of the data measured by the stations already in operation. However, most of the existing monitoring sites are located on high mountains or on the sea shore, while the majority of the area of the continents is of low elevation, close to vegetation and far from the sea.

Inverse modelling techniques (e.g. Enting *et al.*, 1995; Law *et al.*, 1996), among others, combining general circulation models, existing mixing ratio measurements and known source data are used for estimating the location and strength of the “missing sink”. Data series obtained at continental monitoring sites show special features due to local conditions, which have to be taken in account before their inclusion in a global study. The diurnal variation in CO₂ mixing ratios may give information about the spatial representativeness of the data, and may suggest a data selection technique, as will be seen in this paper. The amplitude, relative phase and temporal variation of the seasonal cycle may reflect the location, strength and evolution of the sources and sinks. The average mixing ratio, its trend, and its changes relative to those at other monitoring sites may indicate the areas and factors influencing the global atmospheric carbon budget. This paper presents the 17-year long time series and its characteristic features from a low elevation, continental station located in one of the suspected regions of the “missing sink”, for those, who intend to use the data in global models.

2. Sampling site and instrumentation

K-puszta regional background air pollution monitoring station is located at 46°58'N, 19°33'E, 125 m above mean sea level, on the Hungarian Great Plain, in the middle of the Carpathian Basin (*Fig. 1*). The station is located in a big (approx. 0.3 km²) clearing in a forested area where the immediate surroundings of the station are kept undisturbed by human activity. The nearest public road and settlements (a few hundred inhabitants) are about 3 km to the east/southeast of the station (*Fig. 2*). At the station there is no notable local anthropogenic source (electric heating, no local generator, the staff only consists of a part-time observer). K-puszta is as free from direct pollution as it is possible in highly industrialized, densely populated Central Europe.

The biggest pollution source in this region is Budapest (1.9 million inhabitants), approximately 80 km NNW from K-puszta. The second largest city is Kecskemét (100 thousand inhabitants), located 20 km to the southeast of the station. The prevailing wind in this region is northwesterly and the average wind speed is only 1.9 m/s.

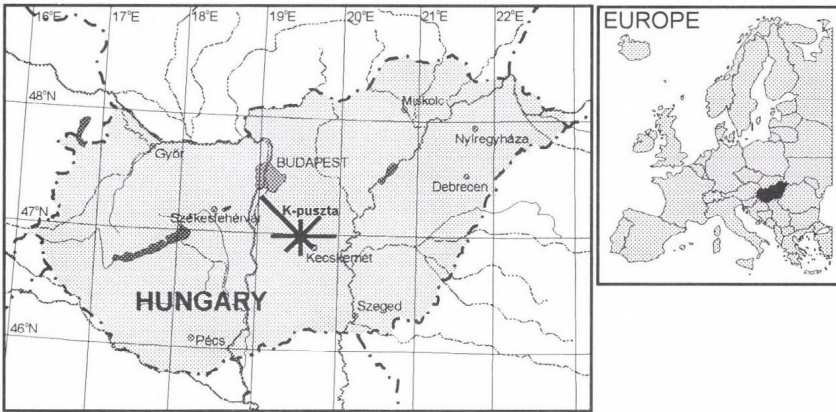


Fig. 1. Geographical location of K-pusztá and major cities (more than 100 thousand inhab.), as well as the frequency distribution of the wind direction at K-pusztá.

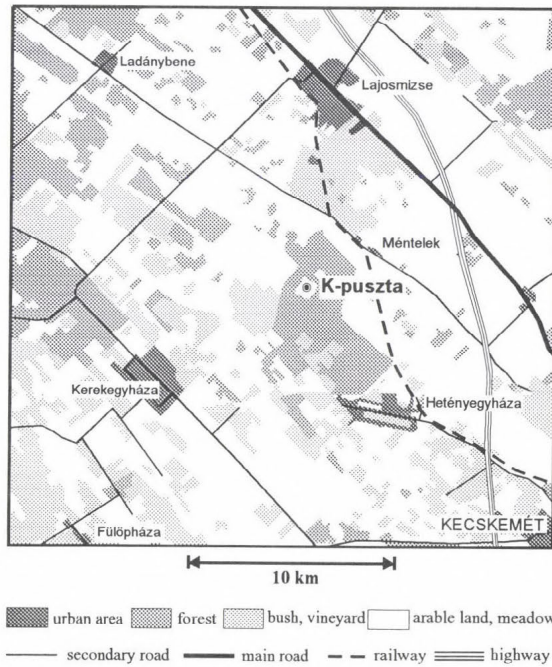


Fig. 2. Region of K-pusztá.

At K-puszta the Institute for Atmospheric Physics of the Hungarian Meteorological Service has been operating a SIEMENS ULTRAMAT 3 CO₂ analyzer since July 1981, for the BAPMoN/GAW program of the World Meteorological Organization. The air inlet of the instrument is at 10 m above grass-covered, sandy ground. Before 1993 the instrument was calibrated against CO₂-in-nitrogen standards provided by the Scripps Institution for Oceanography, U.S.A. (*Haszpra*, 1995). During that time a linear response function was determined on the basis of three standard gases. Since 1993 four CO₂-in-air standards obtained from the National Oceanic and Atmospheric Administration (NOAA-U.S.A.) have been used. For the determination of the mixing ratio quadratic response functions are used. Between two calibrations the response function is linearly interpolated. The pre-1993 data have been corrected against pressure broadening effect *after* the 1992 WMO Round-Robin Inter-laboratory Intercomparison (*Pearman*, 1993). The data presented in this paper are expressed on the WMO Manometric Scale.

3. Data selection and averaging

In order to characterize the carbon dioxide content of the lower troposphere in most of our studies the mixing ratios measured only in the early afternoon hours (12-16 h, local standard time [LST]) are used, when the vertical mixing of the atmosphere is the most intensive (see also Chapter 4.1). For data selection the commonly used clean air sector method (sector from which no anthropogenic influence is expected) cannot be applied because such a sector can hardly be defined: the concentration field is fairly isotropic (*Fig. 3*). It may be the result of the more or less evenly distributed sources and sinks around the station and the large scale mixing in the Carpathian Basin. A similar result was obtained for pollution by anthropogenic non-methane hydrocarbons (*Haszpra et al.*, 1994) which could indicate the direction of the major anthropogenic source areas, like Budapest. As the region is climatologically calm, data selection based on the wind speed (e.g. *Levin et al.*, 1995) is hardly applicable either. Therefore, only the technically false data are rejected. The data are available at the WMO World Data Center for Greenhouse Gases, Tokyo, Japan, and at the Carbon Dioxide Information Analysis Center, Oak Ridge, Tennessee, U.S.A.

The raw data are the 30 minute averages. The 'daily average' mixing ratio of the early afternoon hours is calculated if at least four 30 minute averages are available. The daily average for the whole day is calculated only if 50% of the data is available for both the daytime hours (10-18 h LST) and nighttime hours (21-05 h LST). A monthly average is calculated from the daily averages if at least 10 daily averages are available for the calculation.

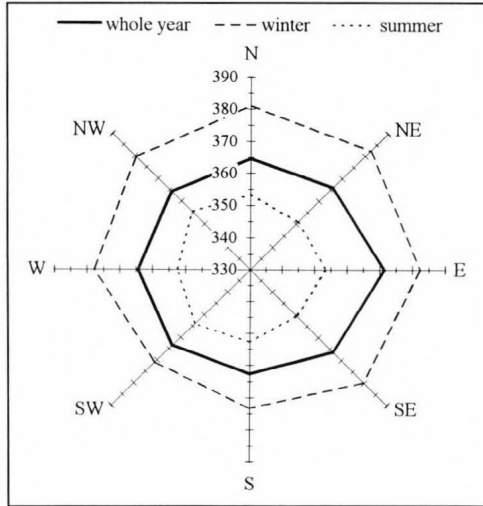


Fig. 3. Sectorial distribution of the atmospheric mixing ratio (ppm) of carbon dioxide at K-pusztá in winter (Dec–Feb), in summer (Jun–Aug) and in the whole year of 1997 in the early afternoon hours (12–16 h LST), when the wind speed is higher than 0.5 m/s. When the wind speed is less than 0.5 m/s the average mixing ratios are 363.7 ppm, 375.4 ppm and 351.8 ppm for the whole year, winter and summer, respectively.

4. Data analysis

Fig. 4 shows the time series of the daily average mixing ratios based on the early afternoon (12–16 h) measurements. The data gaps in the 90's are caused by the long delivery time for spare parts of the instrument and the station reconstruction in 1992, while in the earlier years even obtaining a tank of reference gas was problematic. The average availability of the data between 1989 and 1997 is 72%, including the data gaps in late 1990, 1992 and 1996.

4.1 Diurnal cycle

The station is surrounded by vegetation and located in a flat, low elevation, climatologically calm region, where a shallow nocturnal boundary layer forms frequently. Carbon dioxide emitted by the vegetation and soil accumulates in that layer, where the nighttime mixing ratio may exceed 500 ppm on certain summer nights. The average daily variation relative to the daily average is presented in Fig. 5 for one month in each of the four seasons. The average daily amplitude is as high as 43 ppm in July, while it is negligible (less than 5 ppm) in January. A similar feature can be observed at

other low elevation continental sites like at Waldhof, Germany (53°N, 11°E, 73 m a.s.l.) (Levin *et al.*, 1995). For the interpretation of the diurnal variations of the mixing ratio the average daytime and nighttime depth of the mixed layer are also presented in Fig. 6.

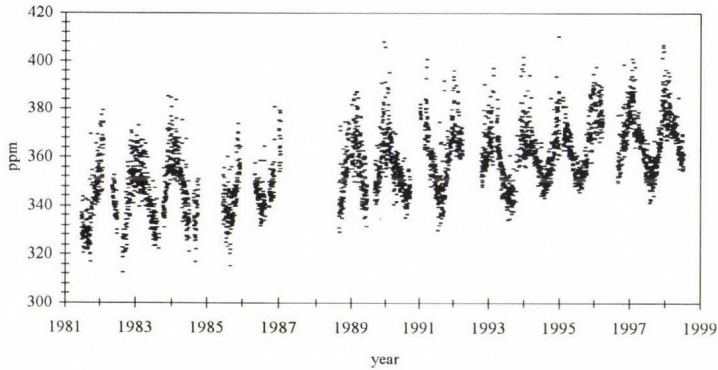


Fig. 4. Daily average atmospheric mixing ratio of carbon dioxide at K-pusztá, based on the early afternoon (12–16 h LST) measurements.

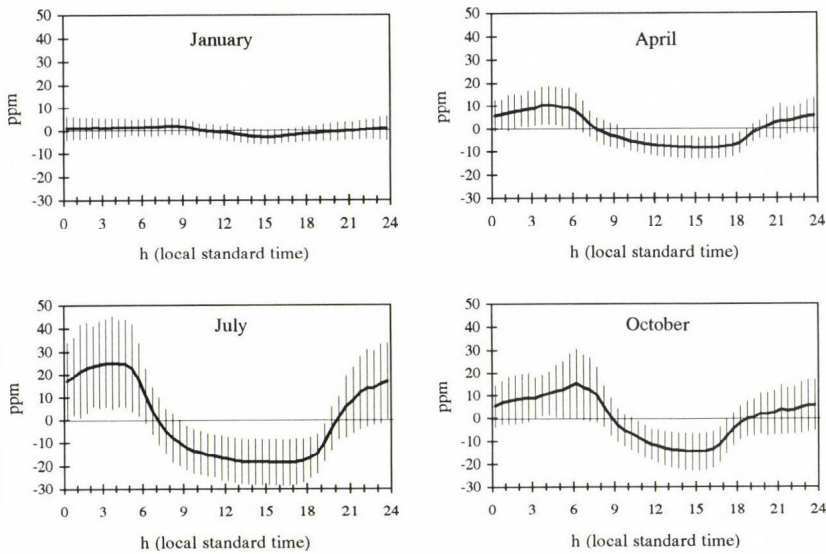


Fig. 5. The average diurnal cycle of CO₂ mixing ratio at K-pusztá relative to the daily average (error bar = $\pm \sigma$).

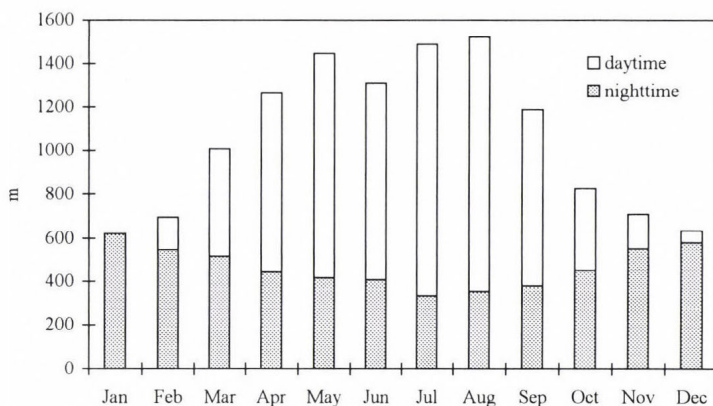


Fig. 6. The average daytime and nighttime depth of the mixed layer between 1987 and 1992 in a suburb of Budapest, 70 km from K-pusztá based on the data and method of Matyasovszky and Weidinger (1998).

The respiration depends strongly on the characteristics of the soil and vegetation, therefore it may be site specific. Consequently, the high nighttime concentrations formed in the shallow stable boundary layer may have much lower spatial representativeness than the early afternoon ones when the mixing processes homogenize a deep layer of the atmosphere. For the regional characterization of the carbon dioxide content of the lower troposphere we use only the data measured between 12 h and 16 h local standard time. The tall tower measurements in the U.S.A. (Bakwin *et al.*, 1995) and the analysis of the data of the German monitoring stations at different elevations (Levin *et al.*, 1995) show that such a data selection criterion may underestimate somewhat CO₂ concentration of the lower troposphere in summer due to CO₂ uptake by the vegetation at the surface, and might overestimate it a bit in winter due to the anthropogenic emission trapped in the shallow boundary layer. More concrete values of the potential bias will be obtained from the Hungarian tower measurements started recently (Haszpra and Nagy, 1997; Haszpra, 1998).

4.2 Seasonal cycle

The mixing ratio measured at K-pusztá shows a pronounced seasonal variation (Fig. 7). The average annual amplitude is about 28 ppm based on the monthly averages. This value would be decreased to 15 ppm if the 'whole-day' values were used for the calculations, mainly due to the increased monthly averages in summer (high nighttime concentrations). The potentially

significant difference should be considered in the analysis of the data measured in the continental region applying different data selection techniques.

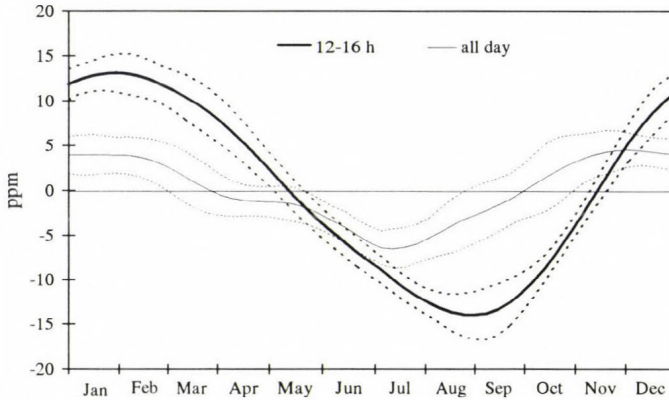


Fig. 7. The average seasonal cycle of CO₂ mixing ratio at K-puszta calculated from the detrended monthly averages (Thoning *et al.*, 1989) with $\pm\sigma$ intervals.

The CO₂ surplus season lasts from early November to mid-May. The minimum concentration is reached at the end of August, while the maximum can be observed in late January. As an additional argument for using only the early afternoon mixing ratio values Fig. 7 also presents the seasonal cycle for the ‘whole-day’ values. This curve shows not only a smaller annual amplitude but also a different shape of the seasonal cycle. It is caused by the special climatology of the Carpathian Basin. In winter cold air may fill up the basin causing limited vertical mixing. The more or less stable air mass breaks up in early spring causing a drop in the concentration that is not attributed to any source or sink.

4.3 Trend

Fig. 8 shows the trend of the atmospheric mixing ratio of carbon dioxide measured at K-puszta. The smoothed curve and the time dependent trend were calculated using the algorithm developed by Thoning *et al.* (1989). From the beginning of the measurements period the mixing ratio has increased from 343 ppm to almost 369 ppm (mid-1998). These values are very similar to the mixing ratios measured at the Northern Hemispheric baseline stations Mauna Loa, Hawaii, and Point Barrow, Alaska, or at the German regional monitoring sites (WDCGG, 1998). If the ‘whole-day’ values were

considered, the corresponding values would be 11 ppm higher at K-pusztá, almost uniformly for the whole period.

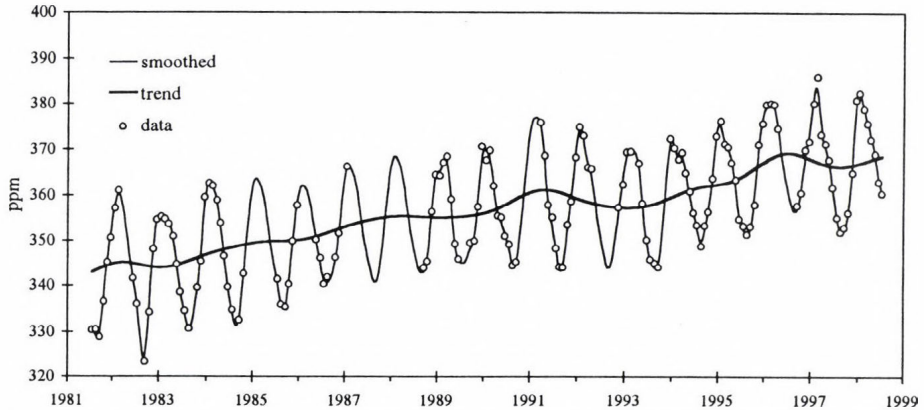


Fig. 8. Trend of atmospheric carbon dioxide mixing ratio at K-pusztá.

The overall trend for the 17 years of the measurements is 1.51 ppm/year. However, similar to the baseline stations mentioned above, the growth rate of the mixing ratio calculated from the time dependent trend was not constant during the period covered. It shows significant fluctuations (Fig. 9). The fluctuation at K-pusztá is more pronounced than at the other stations. At the same time the temporal variation of the growth rates also shows — at least qualitative — similarities. The growth rate is higher than average at each station in 1983, (1985–1986), 1990 and 1993–1995, while it is lower in 1982, (1984–1985) and 1991–1992 (see also Conway *et al.*, 1994). Most of these fluctuations may be triggered by the occurrences of El Niño events in the South Pacific Ocean while the big drop in the growth rate in 1991–1992 might be caused by the climatological consequences of the extreme eruption of Mt. Pinatubo (Lambert *et al.*, 1995). Omitting the period of the long data gap at K-pusztá in 1987–1988 the cross-correlations were calculated for the growth rates observed at K-pusztá, Mauna Loa and Point Barrow as the functions of lag-time (Fig. 10). The maximums of the time dependent cross-correlation of the growth rates are 0.67 for Point Barrow and 0.70 for Mauna Loa, respectively. The high correlation proves that, in spite of the ‘noise’ generated by the nearby vegetation and anthropogenic sources, the global changes are clearly recognizable even in the middle of a continent.

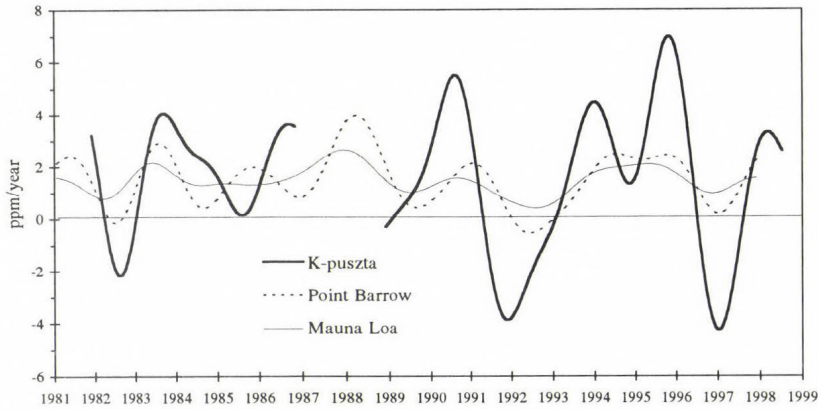


Fig. 9. The growth rate of the atmospheric carbon dioxide mixing ratio.

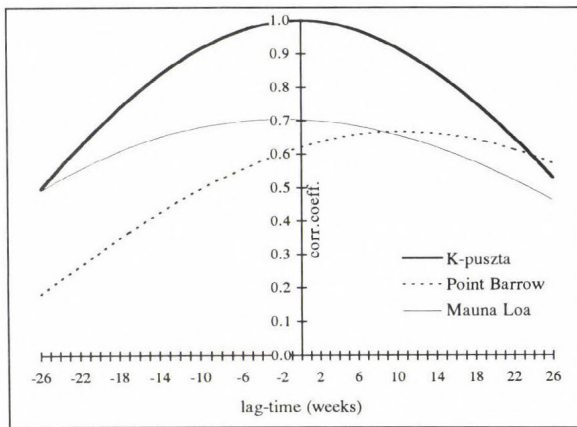


Fig. 10. Cross-correlations of the growth rates as functions of lag-time.

Although El Niño is a phenomenon of the South Pacific Ocean, its climatological effect can also be traced in Europe (*Pongrácz and Bartholy, 1998*). These climatological changes may influence the CO₂ emission/uptake of the vegetation/soil which result in higher fluctuation in the growth rate of the atmospheric CO₂ content over the continent. However, the verification of this hypothesis needs further studies.

5. Summary

The data presented here prove that the global characteristics like the trend, the growth rate fluctuation of the carbon dioxide content of the atmosphere can also be observed at low elevation sites in the middle of a continent. The surrounding vegetation and the climatological calmness of the region results in pronounced diurnal and seasonal cycles of the CO₂ mixing ratio. In this paper we call attention to the importance of the data selection technique at stations under similar environmental conditions that may influence, at least, the average concentrations reported and the characteristics of the seasonal cycle significantly. The "missing sink" question demands continental measurements in spite of their methodological problems. Further research is needed to solve these, usually site specific, problems to get a database suitable for the modeling of the global carbon budget.

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Near surface ozone concentration evaluation and prediction in Budapest

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Abstract—In Budapest near surface ozone levels have been measuring at three points. One of them is representative for the air quality of a large area of the city, for this reason it was chosen for prediction. During the time period (May 1–July 30, 1997) the meteorological parameters and ozone levels were examined as well. It is found, that temperature and wind speed have an influence on ozone values. An increase by leaps and bounds of temperature causes an increase of ozone concentration. A combination model consisting of a deterministic (OZIPR) and a statistical model were used for forecasting the ozone maximum the next 10 hours. To improve the results coming from the photochemical model a regression analysis was applied which was based on meteorological condition and emissions in Budapest. The verification of the forecast shows that the obtained results are better than using a persistency model.

Key-words: urban ozone concentration, forecasting system, photochemical transport model, multiply regression analysis, model evaluation.

1. Introduction

At the Ministerial conference in London, May 1996 a Technical Working Group (TWG) on Data Exchange and Forecasting for Ozone Episodes was formed to develop a co-ordinated system for data exchange and forecasting air pollution episodes in Europe. The exchanged data are made directly available to the public, for instance via a world wide web page, to emphasise the transboundary nature of ozone episodes in Europe.

The Working Group concluded that a central system operating at the regional scale is needed. A joint European forecast system will provide information on larger spatial scales which will improve the insights during the build-up and further development of actual ozone episodes. The results of

such a causal model will form essential inputs to national systems. It is judged that national models, in particular statistical models, based on local conditions and local inputs will have better performance than a European-scale model. Consequently, using a national ozone forecasting model is indispensable to an adequate ozone prediction and for extending the forecast period to 2 or more days. For this purpose, a Hungarian ozone forecasting system has been under development. Moreover, when the current national forecast models are used in combination with the results of a large-scale photochemical model, better performance might be obtained.

Near surface ozone is commonly perceived to be a regional pollutant. For this reason in many European countries ozone prediction models are used on the regional scale. However, smog problems can arise in big cities like Budapest where the density of population is quite high.

In this paper a smog forecasting system has been used for Budapest to predict the maximum ozone concentration for the next 10 hours. This system consists of a deterministic and a statistical model. The deterministic model is the photochemical transport model developed for the U.S. Environmental Protection Agency (EPA) called OZIPR, OZIPR (R for research) is a research-oriented version of EPA's OZIPP (Ozone Isopleth Plotting Package) computer modeling program. This air quality simulation model, in conjunction with the Empirical Kinetics Modeling Approach (EKMA), relates ozone concentration to levels of organic and nitrogen oxide (NO_x) emissions. A statistical model has been developed for emissions and meteorological conditions in Budapest. This forecasting system was tested here for the summer period in 1997.

2. Data evaluation for 1997's summer period

Spatial variations in O_3 concentrations occur on the macro- and microscales, as well. These variations are relevant for exposure assessments and for understanding and controlling the photochemical formation of O_3 . Ozone concentrations differ between urban and rural areas, between urban and remote sites, and even between rural and remote sites, as a function of space and travel time of plumes, in addition to other factors. For exposure assessments, however, it is important to note that fairly significant intra-city and intra-airshed variations in O_3 concentrations can occur.

O_3 concentrations are often higher in some suburban or even rural areas downwind of urban areas than in the urban areas themselves. The time required for the development of photochemical reactions, transport of the urban plume, and the absence of fresh NO emissions as the plume travels, are key factors in producing higher concentrations in some downwind areas.

There are three ozone measurement sites in Budapest. The first one (Laborc Street) is situated in the north-west part of the city, the second one (Baross Square) is in the downtown area (these ozone values are lowest), and the third ("Fodor József" National Centre of Public Health, National Institute of Environmental Health, NIEH) is located in south-east part of Budapest. The prevailing wind direction is from north-west, and these stations are located along this direction crossing the town. Daily average ozone data coming from these stations have been compared to the regional background concentrations, which are measured at the K-pusztá station 70 kms from Budapest (in the east-south direction), as *Fig. 1* shows. The data covers the period from March to July in 1997, when relatively high ozone concentrations were measured. In August measurements were not available in Budapest from the NIEH station so that only three months of 1997' summer period ozone data were examined at the urban and in rural sites. The highest values can occur at K-pusztá station and an appreciable intracity gradient in O_3 concentrations can develop across the city. For the prediction of ozone concentration the NIEH data were chosen, because this monitoring site is located in the downwind area of the city, and its location is quite high (20 m above ground-level) so higher values can occur.

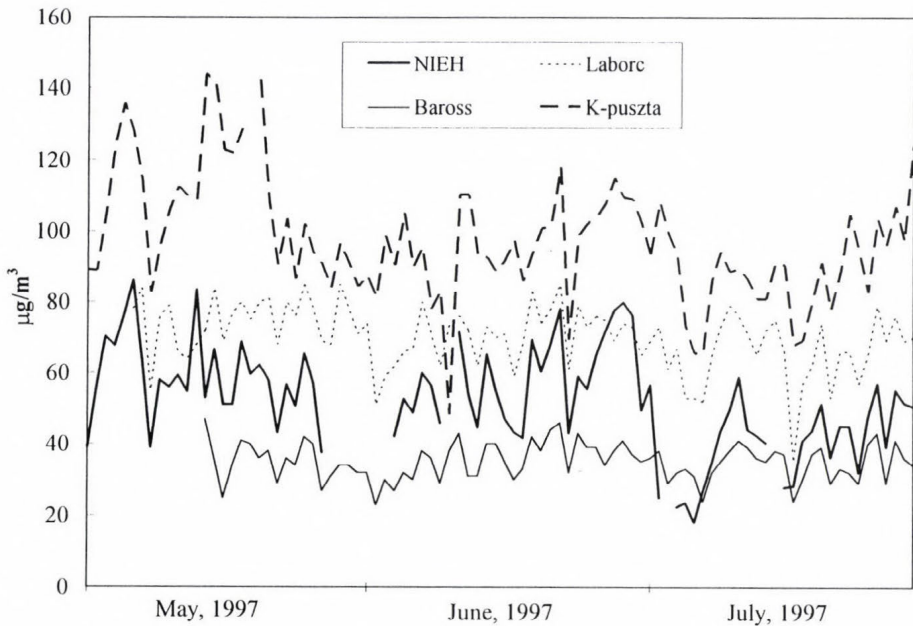


Fig. 1. Daily average ozone concentrations at four monitoring sites.

It can be seen in Fig. 1 that ozone episodes are not only a local, but a regional phenomenon. At the beginning of May, and at the end of June the ozone level was elevated at K-puszta, and at the same time high values were detected at the NIEH station.

The development of ozone concentrations at the NIEH station is displayed in two time scales in Fig. 2. This allows the diurnal variation and the development of an ozone episode over several days to be shown independently. Elevated concentrations occur, in general, between 10 a.m. and 8 p.m. Ozone levels higher than $100 \mu\text{g}/\text{m}^3$ can be observed at the beginning and at middle of May, and at beginning and at the end of June. A temporary decrease can be seen in July.

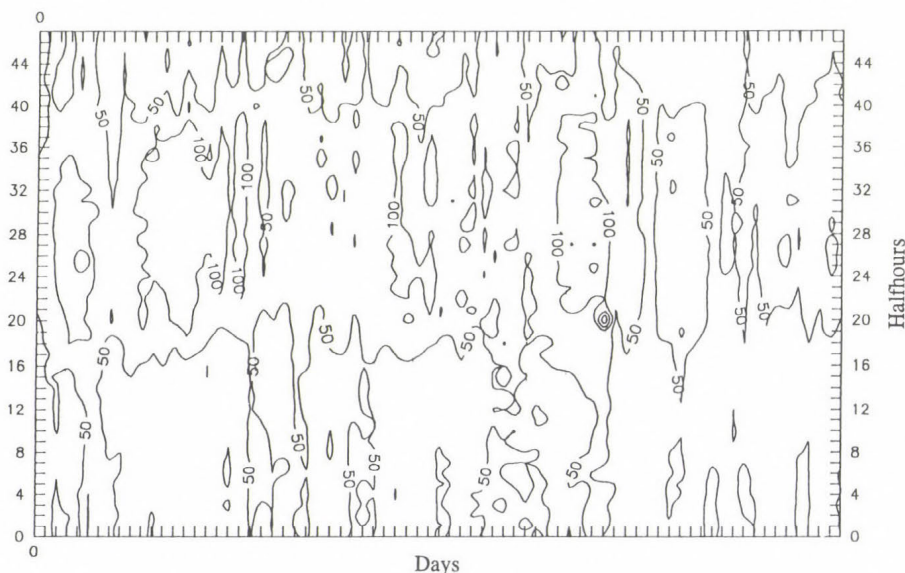


Fig. 2. Half-hour mean values of ozone concentration observed in Budapest during the time period from 1 May to 31 July, 1997.

3. Meteorological conditions

When the ozone concentration was high, during the time period from 1 May to 30 June, 1997 hourly values of ozone concentration, air temperature, wind velocity and wind direction have been monitored in Budapest. Fig. 3 a-c. shows that NW and SE directions are preferred in frequency, and NW winds are the strongest. These strong NW winds may represent the weather fronts

of an anticyclone passing from NW to SE. Interestingly, the highest concentrations in ozone are associated with westerly winds. This fact can be explained by the position of the monitoring station that is situated in the eastern part of Budapest and is affected by the urban plume of the city.

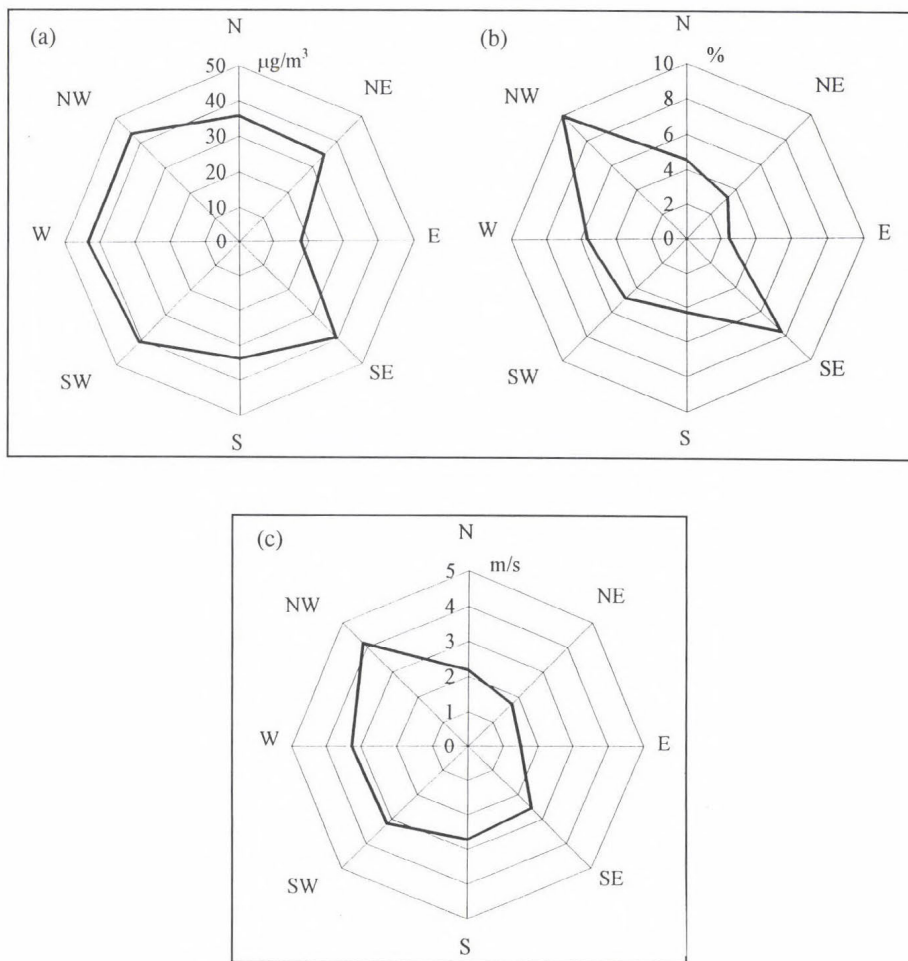
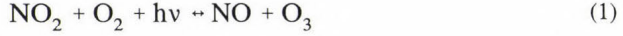


Fig. 3. (a) Windrose for ozone concentration, (b) wind frequency and (c) wind velocity.

Tropospheric ozone is formed in photochemical reactions. The chemical equilibrium of



is disturbed by the presence of CO and/or non-methane hydrocarbons. Such substances 'catch' NO and, thus, shift the equilibrium Eq. (1) towards ozone production (Graedel and Crutzen, 1993). In this manner, the concentration of precursors and the strength of the solar radiation determine the amount of the ozone that is produced. Besides this, the temperature influences the efficiency of the reaction (1). Apart from air-mass convection, air temperature is an indicator for the strength of solar radiation.

Ozone concentration and air temperature are displayed simultaneously in Fig. 4. The peaks temperature and ozone concentration coincide, which means that the ozone concentration increases with temperature. Generally, the ozone concentration depends on the intensity of the UV radiation. Often the rise of the temperature coincides with a rise of UV-radiation. It follows, that ozone concentration should be correlated with air temperature. This way, the hypothesis that ozone concentration is associated with changes in temperature could be established here.

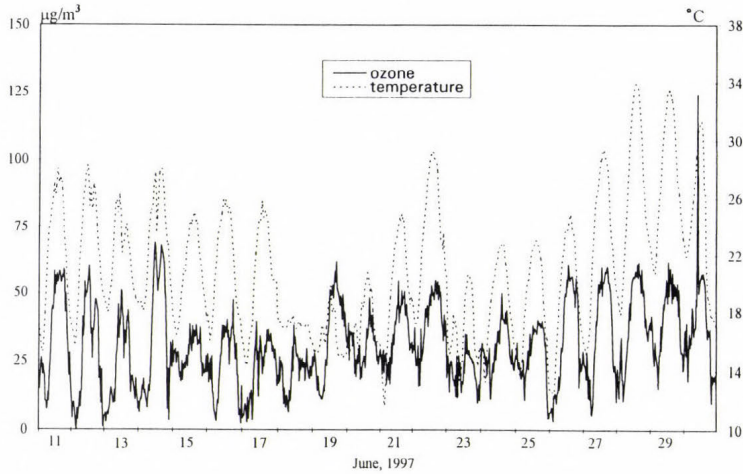


Fig. 4. Ozone concentration (solid line) and temperature (dashed line).

In order to study the impact of air movement on ozone accumulation, ozone concentration is plotted parallel to observed wind speed and direction in Fig. 5 and 6. Fig. 5 compares the ozone concentration with the wind speed. It is possible to see an association between both variables as the plots

of wind speed looks nearly similar to the plot of ozone concentration. Often the peaks occur at the same time. Sometimes there are also exceptions, for instance on 14. 06. 1997 about midday where the wind speed is relatively low and the ozone concentration is high.

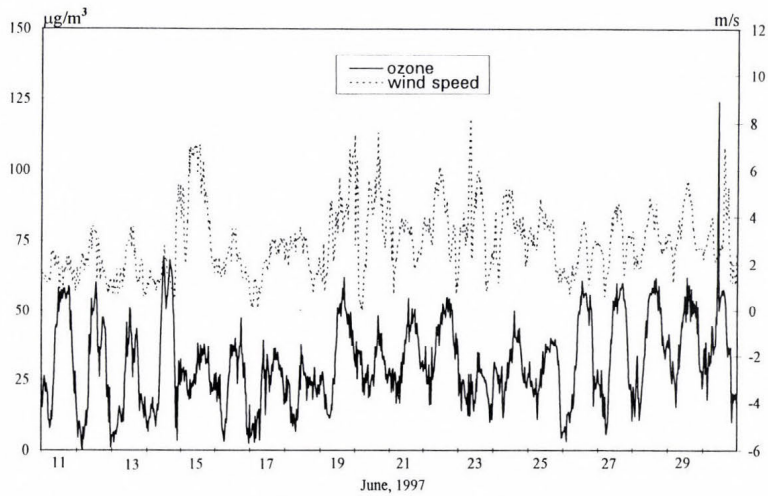


Fig. 5. Ozone concentration (solid line, left axis) compared with wind speed (dashed line).

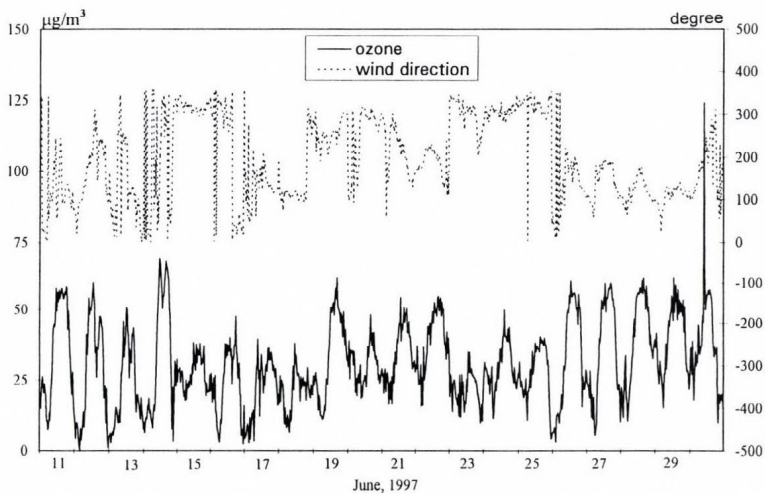


Fig. 6. Ozone concentration (solid line, left axis) compared with wind direction (dashed line).

In Fig. 6 the ozone concentration and direction of wind are not synchronous, that means that the times of peaks do not coincide and that the plot of wind direction has a structure different from the plot of ozone concentration.

In summary, it is found that temperature and wind speed have an influence on ozone concentration. It also seems that the direction of wind has no influence on ozone concentration, but this conclusion must be studied in more detail.

4. Forecasting

Episodes of photochemical air pollution are periods of some days to some weeks with calm, warm, sunny weather, in which atmospheric photochemical reactions of nitrogen oxides and volatile organic compounds lead to a host of mostly noxious air pollutants. Ozone is considered to be the most important of these in view of its concentrations and its effects on human health.

A hybrid model for the prediction of the ozone episodes at one point, the NIEH station, was developed. This system consists of a deterministic and two statistical models. In the deterministic model, the concentration is calculated from emissions of ozone precursors (volatile organic compounds and nitrogen oxides) and forecasted meteorological conditions in Budapest taking into account relevant atmospheric processes such as dispersion, transport, chemical conversion and deposition. The statistical model is based on a multiple linear regression analysis.

4.1 Deterministic model

OZIPR a new trajectory-type, air quality simulation model based on EPA's Ozone Isopleth Plotting Program (OZIP) Model (*Gery and Crouse, 1996*), which was established for regulatory purposes but contains improved and expanded capabilities that make the model useful for research purposes. OZIPR serves the dual purpose of providing: (1) a simple trajectory model capable of utilizing complex chemical mechanisms, emissions, and various meteorological parameters, (2) procedures through which the Empirical Kinetics Modeling Approach (EKMA) can be implemented for calculation of emission reductions needed to achieve compliance with the National Ambient Air Quality Standard for ozone. While these capabilities were included in earlier versions of OZIP, the new OZIPR contains major improvements that increase the program's ability to input, utilize, and output a much larger range of information.

The OZIPR simulates complex Chemical and Physical processes of the lower atmosphere through use of a trajectory model. The model incorporates the Carbon Bond 4.0 (CBM-IV) chemical mechanism (*Whitten and Gery,*

1986) which are complex chemical mechanisms used to describe the chemical processes that occur within the modelled air mass. The chemical mechanism in the model is based on the Carbon Bond Mechanism IV (CBM-IV) and contains 37 compounds and about 80 chemical reactions for the numerical solution of the chemistry and the application of the chemical mechanism). The physical representation is a well-mixed column of air extending from the ground to the top of the mixed layer. So the determination of the mixing height is a very important parameter. This idealized air column moves with the wind (along the wind trajectory, but cannot expand horizontally). Emissions from the surface are included as the air column passes over different emission sources, and air from above the column is mixed in as the inversion height rises during the day.

OZIPR model input parameters along the trajectories are: co-ordinates; time zone; date of the day (year, month, day), the starting hour and the end of the simulation. The model required meteorological and air quality inputs are shown in *Table 1*.

Table 1. Meteorological and air quality inputs of OZIPR model

Meteorological inputs	Predicted values	Measured values	Average values
Height of mixing layer at 8 a.m.			✓
Maximum height of mixing layer		✓	
Surface temperature	✓		
Relative humidity		✓	
Atmospheric pressure		✓	
Dry deposition velocity			✓
<hr/>			
Air quality inputs			
O ₃ , NO _x , VOC and CO concentrations at 8 a.m.		✓	
Aloft of O ₃ , NO _x , VOC and CO con.			✓
NO ₂ /NO _x ratio at 8 a.m.		✓	
Reactivity of VOC			✓

The height of mixing layer is one of the fundamental parameters to characterize its structure and is required in dispersion models. Note that the maximum values of the mixing height are determined by midnight radiosonde measurements using Holtzworth's method. The other essential input para-

meters are emission values estimated from values in U.S. cities and adapted to the sources condition of Budapest.

OZIPR model provides a 10-hour ozone forecast each morning at 8 a.m., that is to say the expected maximum value of the ozone concentration of the afternoon for one ozone monitoring site in Budapest.

4.2 Statistical models

Most of the national ozone forecasting systems use statistical (multiple regression) models. Temperature and previous day's ozone levels are commonly used as independent variable. In more recently developed systems, neural networks are frequently used. Output of the system is tomorrow's maximum ozone level, that is a forecast span of less than 24h; forecasts for 48h are given by one or two systems only.

We have established a statistical model for improvement of the deterministic model results. This calculation is based on a multiple linear regression involving only one ozone measuring site (NIEH) in Budapest. The maximum ozone concentration for 10 hours ahead is the output. Temperature and the previous day's ozone levels are used as independent variables.

The maximum ozone concentration for the actual day is calculated by using the max. O_3 concentration coming from the OZIPR model (max O_3 model), the temperature maximum for the actual day coming from weather forecasting (max Tact), as well as the maximum ozone concentration of the previous day (max O_3 prev), and the temperature maximum of the previous day (max T prev). The two latest independent variables are based on measured data. These inputs are coupled by the following equation are provide the maximum ozone concentration prediction for the next 10 hours:

$$\begin{aligned} \max O_3 \text{ prog} = k + a (\max O_3 \text{ model}) + b (\max Tact) + \\ c (\max O_3 \text{ prev}) + d (\max T \text{ prev}), \end{aligned} \quad (2)$$

where k , a , b , c , d are regression coefficients. Statistical parameters of the regression model are summarized in the *Table 2*.

Using this method we can obtain more adequate results than when only running a deterministic model, but extreme events (situation far from averages) can not be predicted well. This phenomenon is a feature of the statistical model. If not only the temperature, but other meteorological parameters which have an influence on ozone formation, are included in the model, more reliable results can be obtained. Moreover the long-range transport, which affects ozone levels has not been taken into account.

Table 2. Statistical parameters of the linear regression model

Abbreviation	Definition	Values
k	The constant of the linear regression	-405.567
a	The regression coefficient of the first independent variable	0.229
b	The regression coefficient of the second independent variable	0.680
c	The regression coefficient of the third independent variable	-0.050
d	The regression coefficient of the fourth independent variable	0.826
se _k	The standard error value for the constant k	210.952
se _a	The standard error value for the coefficient a	0.225
se _b	The standard error value for the coefficient b	0.877
se _c	The standard error value for the coefficient c	0.129
se _d	The standard error value for the coefficient d	0.852
r ²	The coefficient of determination	0.119
se _y	The standard error for the y estimate	19.741
F	The F statistic	2.326
df	The degrees of freedom	69.000
SS _{reg}	The regression sum of squares	3625.621
SS _{resid}	The residual sum of squares	26890.080

5. Evaluation of the results

Air-quality models are evaluated by comparing their predictions with ambient observations. For validation of the forecast procedure independent data, that is, data not used in the development of the (statistical) model is needed, and it is desirable to have several years of data available for evaluating model performance. If this is not feasible an alternative procedure might be followed, in which skill parameters are introduced.

For quality information, the skill of forecast system can be tested by using a binary system for comparison of forecast and observed occurrence of air quality classes, that is using a standard contingency table of the type displayed in Table 3, where N is the total number of data points and defining: f = total number of forecast exceedances, m = total number of observed exceedances, a = number of correctly forecast exceedances. The other matrix elements in Table 3 can be expressed in N, m, f, and a. This method are proposed by TWG (*van Aalst and de Leeuw, 1997*) as well.

Table 3. Contingency table used for verification of ozone forecast

Forecast	Observed		Total
	Yes	No	
Yes	a	f - a	f
No	m - a	N + a - m - f	N - f
Total	m	N - m	N

For this kind of evaluation the ozone threshold values are needed. Nowadays the following threshold values are valid in Budapest: for 24 h it is $100 \mu\text{g}/\text{m}^3$, for one half-hour it is $110 \mu\text{g}/\text{m}^3$. A smog event is defined as a day with maximum one-half-hour ozone concentration exceeding $110 \mu\text{g}/\text{m}^3$.

Using these definitions, various skill parameters can be defined:

- fraction of correct forecast smog events (probability of detection)

$$SP = a/m \cdot 100\% . \quad (5)$$

Note that the fraction of “unexpected events” is given by $(100 - SP)\%$;

- fraction of actual forecast smog events

$$SR = a/f \cdot 100\% . \quad (6)$$

Note that the fraction of “false alarm” is given by $(100 - SR)\%$. Both SP and SR range from 0 to 100 with a best value of 100.

In an overall assessment of the forecast skill for all four matrix elements have to be considered. The weighting of each of the four elements in the total skill might depend on the objectives of the forecast system. When the main objective is the warning of sensitive population groups the number of unexpected smog events should be minimal. In cases where the forecast is used as a trigger for short-term abatement measures (e.g., traffic ban) the “false alarms” should be minimal in view of the high costs involved in taking this kind of measures. A high false alarm rate will also reduce the confidence of the general public in the system.

- Assuming an equal weight to the correct forecasting of smog events and of non-smog events, the scoring parameters SP and SR can be combined to form a success index, SI, ranging from -100 to 100 with a best value of 100 :

$$SI = \left(\frac{a}{m} + \frac{N + a - m - f}{N - m} - 1 \right) \cdot 100\% . \quad (7)$$

For evaluating of the numerical output of a forecast (sub)-system the following skill parameters, each evaluating different aspects of the system, can be considered:

- the fractional bias, FB, between averaged values for forecast and observation

$$FB = 2 \frac{(\bar{P} - \bar{M})}{(\bar{P} + \bar{M})}, \quad (8)$$

where P is the forecast value and M is the observed value. FB will detect any systematic difference between observation and forecast. FB ranges between -2 and $+2$ with the best value of 0 ;

— the skill score, S defined as:

$$S = 100 \left\{ 1 - \frac{\sum (P_{i+1} - M_{i+1})^2}{\sum (M_i - M_{i+1})^2} \right\}, \quad (9)$$

where P_i and M_i are forecast, respectively observed values at day i . The score S includes the persistency model as reference model. A value $S < 0$ indicates that the forecast model is worse than the persistency model. A score $S = 100$ indicates the perfect model;

— the hit score, H , can be defined as:

$$H = 100 \frac{1}{N} \sum \frac{2r - \Delta}{2r}, \quad (10)$$

where $\Delta = \min(|P - M|, 2r)$ and r is an uncertainty range around the forecast and observed value. The uncertainty in the observed value will depend on, e.g. uncertainties in the measurements but more importantly on the spatial variability of M within the study area. The uncertainties in forecast value will result from the model concept. When these uncertainties are not known, the hit score may provide a simple estimate of their impact on forecast performance. The hit score H indicates the overlap between the interval $(P \pm r)$ and $(M \pm r)$. H ranges between 0 and 100 with a best value of 100 . Disadvantage of H is that it assumes a uniform distribution of P , M within the interval $\pm r$ and it assumes equal ranges around both P and M . The hit score, H , has been evaluated with a range $r = 10 \mu\text{g}/\text{m}^3$.

In the evaluation of skill parameters a reference model should be included. The persistency model is recommended as the reference model. The persistency model is an extremely simple model assuming that the forecast situation equals the present situation. In view of the phenomenology and average duration of ozone smog episodes the persistency model will be reasonably successful; the skill of the forecast system should be better. In practice this means that the forecast should be able to predict abrupt changes in air quality levels.

When skill parameters are all scaled between 0 and 100 summarising graphs like Fig. 7 can be made. Using such a figure, a direct comparison between the forecast model and the reference (persistence) model can be made. The figure and Table 4 show that the developed hybrid model for 10-hour ozone prediction provides better results than the persistence model. So it can be used operationally.

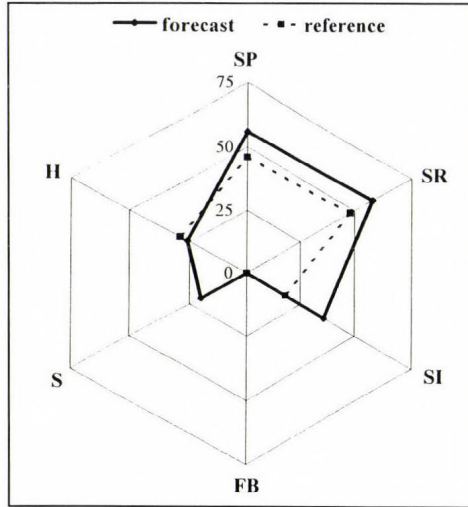


Fig. 7. Summary of evaluation of the ozone forecast system for Budapest in summer 1997.

Table 4. Evaluation of the ozone forecast of summer 1997 (May 1–July 31) made for Budapest

Forecast	Observed		
	Yes	No	Total
Yes	14	10	24
No	11	39	50
Total	25	49	74

Number	Skill parameters	Ozone forecast	Persistency model
1	SP	56	46
2	SR	58	48
3	SI	36	18
4	FB	0	0
5	S	20	n.a.
6	H	26	29

The prediction of ozone concentration is not so easy because oxidant formation seemed to be very episodic, and episodes appear to be of two types: local/mesoscale and transport (*Schjoldager*, 1981). This approach does not consider the mesoscale and long-range transport episodes. The aim of the future research is to extend the output for the whole area of Budapest, and to place uncertainties on the output values. The updating of emission data will be investigated as well.

6. Conclusions

In summary, the following conclusions are drawn:

- Ozone can accumulate when there are high temperatures and stagnant air in Budapest.
- A trajectory-type ozone simulation model and a statistical model were used to predict the expected maximum concentration of the next 10 hours for one monitoring site of Budapest.
- Advantages of forecasting system are ease of use due to small number of predicted input parameters, and ability to deal with transport phenomena when the changes of local precursor emissions are known.
- Better results are obtained by using the forecast model than by relying on persistency.

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The effect of alginite on the production and water use of maize

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Abstract—Bioproduction and environment-friendly agriculture need non-toxic, natural materials to improve soil conditions for maintaining the production on a reasonable level. Alginite is originated from the Pliocene age located at the site of the past Pannon Sea. The aim of our investigation was to determine the effect of alginite on the production level and water use of different maize hybrids as well as the mechanism of its influence. The hybrids were Norma 370 SC a water stress tolerant hybrid and MVK-480 SC which was bred for irrigated conditions. Experiments were carried out in 1996 and 1997 at Keszthely, Hungary (46°46'N, 17°14'E, 128 m a.s.l.). Treatments were as follows: (1) C: Control (without alginite and nitrogen fertiliser); (2) A: Alginite 5 kg m⁻² (8 g kg⁻¹ soil), no nitrogen fertiliser; (3) N: No alginite, 300 kg ha⁻¹ fertiliser (10 g m⁻² N, 8 g m⁻² P, 12 g m⁻² K fertilisers); (4) A+N: Alginite 5 kg m⁻² (8 g kg⁻¹ soil), 300 kg ha⁻¹ fertiliser (10 g m⁻² N, 8 g m⁻² P, 12 g m⁻² K fertilisers). The plants were grown in evapotranspiration pots 4 m² each in three replications. The responses of hybrids to alginite were slightly different. Leaf area increase due to alginite was more expressive in the case of Norma than in MVK-480 especially in treatments without nitrogen application. Alginite caused significant depression in water use in the case of cv. MVK-480 comparing treatment A and C as well as in treatments A+N and N. In the case of cv. Norma differences were not significant because of the leaf area response. In the beginning of the growing season an increased stomatal resistance could be measured in the alginite treatments in accordance with decreased water loss at that time. The effect of alginite as a nutrient is more expressive without nitrogen fertilisation. In 1996 there was 7% and 10% more yield in treatments A comparing to control C in MVK-480 and Norma, respectively. In the second year, 1997 the effect was stronger (30% and 74%). Reason of difference between the individual treatments might have been on the one hand the nutrient effect of alginite on crop production and on the other hand the change in soil hydrophysics, that could result a humid-warm microenvironment which has a feedback to the evapo-transpiration processes.

Key-words: alginite, evapotranspiration, micro environment, plant production.

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1. Introduction

Agriculture is often blamed for contaminating land with chemicals. The importance of environment friend farming has been growing fast in past decades. Searching for materials which are suitable from this point of view began in the eighties. The alginite as a substitute of artificial fertilisers satisfies the most rigorous laws and requirements of bioproduction technologies avoiding the excess use of chemicals. For this reason it is an agent much in demand, especially on areas where environmental control has top priority. Alginite is the local resource of strictly protected region, the Lake Balaton catchment area, where the investigations had been carried out and this natural material would be applied. The most important advantages of alginite are decreasing nutrient infiltration and lake eutrophysation. For this reason we could not give up its use in our surroundings, mainly in the closest fields to the lake.

After the volcanism of Pliocene time, the Pannonian lakewater infiltrated through the pores and fissures of the tuff-ring and formed a deepwater (50 to 120 m) crater lake extremely enriched with nutrients (Nagy, 1978; Hargitai, 1985). The wealth of mineral and organic nutrients led to a marked growth of green algae (*Botryococcus braunii*). When dead, they would settle at the lake-bottom to accumulate as an alginite bed, that may be taken to be a fossil biomass (Knutson *et al.*, 1986). Superimposed on an organic rich layer, a thin layer of carbonate rich sediments (lime, dolomite, diatomite) was laid down in an arid growing season. Process was repeated several times producing pairs of laminae composed of algae on the one end and clay, lime and diatoms on the other one (Ravasz and Solti, 1987). This mixed material is called alginite. Because of its special composition, it is suitable for complex soil melioration, for the structure of the soil, its water and nutrient regime and humus content can be simultaneously improved (Russel, 1992). Due to the composition of alginite, huge amount of literature introduced it as a substitute of fertilisers (Solti, 1985, 1985a, 1987; Solti and Szabó, 1985; Solti *et al.*, 1985). Chemical compounds of the alginite are presented in Table 1.

Table 1. Chemical compounds in g kg⁻¹ of alginite of Hungarian origin

N Total	N Hydrolysable	P	K	Ca	Mg	Organic matter
3-5	0.12-0.2	5-6	6-9	100-300	8-10	200-300

In spite of the relatively high nitrogen, phosphorous and potassium (NPK) content of alginite, the soluble mineral rate of macroelements is low. In case of N the ratio of hydrolysable N is only 4%. Therefore the alginite can not be taken as a complete nutrient source. Size of yield increase depends on applied

dose. Beside that alginite may be a crop nutrient supplement, its high water retaining capacity has of primary importance in dry seasons (*Russel, 1992*).

Our experimental field is located on highly protected recreational area of Hungary, on Lake Balaton catchment area where decrease of nutrient leaking into the ground- and lake water is very important. Since alginite is a non toxic environment friend natural material, satisfying the most rigorous environmental laws and restrictions, it could be applied in crop growing of bioproduction. The alginite is a local resource of Lake Balaton region, therefore transportation expenses are low when it is used close to the mine.

The aim of our investigations was to determine the changes in yield and water regime of two different maize hybrids due to alginite treatment. Possible explanation of alterations in plant and environmental factors resulted from alginite application are discussed as well.

2. Materials and methods

At the Agrometeorological Research Station of Keszthely, Hungary there are 24 Thornthwaite type compensation evapotranspirometers (ET-pots) with a surface of 4 m² and a depth of 1 m each. This apparatus provides unlimited water supply from below the pots. Water consumption of the soil-plant system can be measured on a daily basis. Actual evapotranspiration is calculated using the values of water uptake and potential evapotranspiration. In this study we used to estimate the daily values of potential evapotranspiration according to local formula of *Antal* (1968). The soil in the pots is a Ramann type brown forest soil with mean bulk density of 1.5 Mg m⁻³ in the top 1 m of the profile and a field capacity of 290 mm m⁻¹. The pH of the soil and alginite were 6.3 and 6.9, respectively.

Experiments were conducted in 1996 and 1997. Maize hybrids were studied; namely Norma 370 SC which is a water stress tolerant hybrid and MVK-480 SC which was bred for irrigated conditions. Both of them are commercial varieties in Hungary. Treatments were as follows:

- (1) C: Control – without alginite and nitrogen fertiliser.
- (2) A: Alginite 5 kg m⁻² (8 g kg⁻¹ soil), no nitrogen fertiliser.
- (3) N: No alginite, 300 kg ha⁻¹ fertiliser (10 g m⁻² N, 8 g m⁻² P, 12 g m⁻² K fertilisers).
- (4) A+N: Alginite 5 kg m⁻² (8 g kg⁻¹ soil), 300 kg ha⁻¹ fertiliser (10 g m⁻² N, 8 g m⁻² P, 12 g m⁻² K fertilisers).

A randomised complete block design with three repetitions was used. Alginite and fertiliser were incorporated in the upper 40 cm soil layer at the end of April, 1996, just before planting. Seeds were sown on 30th of April, 1996 and 15th of May, 1997. The plants emerged on 12nd and 29th of May,

respectively. There were 28 plants in each ET-pot, i.e., a plant density of 7 plants m⁻², or 70 000 plants/ha. The pots were surrounded with maize having the same density.

Assimilatory surface was characterised by leaf area index (LAI) measured weekly using LI-3000 type portable leaf area meter (LI-COR, Lincoln, NE). Leaf area of the same 10 plants per pot was measured during the entire growing season.

On a few sample days yearly, air temperature and relative humidity within the canopy were registered at three levels (ground-, cob-, and top levels of plants) with psychrometers connected to a LI-COR Model LI 1000-32 datalogger. The sensors were shaded to remove the effect of direct radiation.

Soil temperature was sensed remotely on the same days when stomatal resistance was also measured. An infrared thermometer of Raynger II Model (Raytek, Santa Cruz, CA) was used with a 20 field of view and an 8–14 μm spectral band filter. Emissivity of soil was assumed to be 0.9 (Anda, 1993). Average soil surface temperature was measured by walking along the rows for 60 seconds holding the instrument at 0.1 m, just above the soil surface.

At the end of the growing season, cobs were harvested. Fresh weight was measured and a sample of three cobs per pot was oven-dried at 60°C for 48 hours to obtain moisture content of the cobs. Data were analyzed using combined analysis of variance across the years.

3. Results and discussion

3.1 Weather of the experimental seasons

The growing season 1996 began with a warm and rainy period (Table 2). It was favorable for germination and early development of the plants. A longer dry period started at the end of May and continued in June. The temperature in the middle of June was above the long term average. The second dry period occurred between July and the 2nd pentad of August. During this time temperatures were rather cool. September was extremely wet and cold. The spring of 1997 was very dry until the 4th pentad of May. Heavy rains provided sufficient water for development until the end of July. August and September were dry again with rather cool temperature.

Table 2. Monthly mean temperature (T, °C) and amount of precipitation (P, mm) in the growing season of 1996 and 1997 at Keszthely, Hungary

		Apr	May	Jun	Jul	Aug	Sept
1996	T	10.1	16.4	19.7	18.9	19.5	12.3
	P	84.0	79.1	38.2	56.8	82.6	171.6
1997	T	7.7	16.6	19.3	19.5	19.9	14.8
	P	10.9	75.0	93.7	111.0	27.2	21.5

3.2 Assimilatory surface (LAI)

Seasonal variation of leaf area index is shown in *Fig. 1a* and *1b*. Maximum leaf area was reached middle of July 1996 and early August 1997, respectively. Both alginite and nitrogen caused some increase in LAI. Compared to treatment C, treatment A had a 30–50% ($P < 0.01$) greater LAI in the case of cv. Norma and a 5–13% ($P < 0.05$) increase occurred in the case of MVK-480. The well known nitrogen effect on LAI was also demonstrated in our experiments. When alginite was applied together with nitrogen, the differences in LAI between the treatments A+N and N were not significant for both cultivars.

Senescence of leaves in plant stands grown without N was always faster than in canopies with additional N fertiliser.

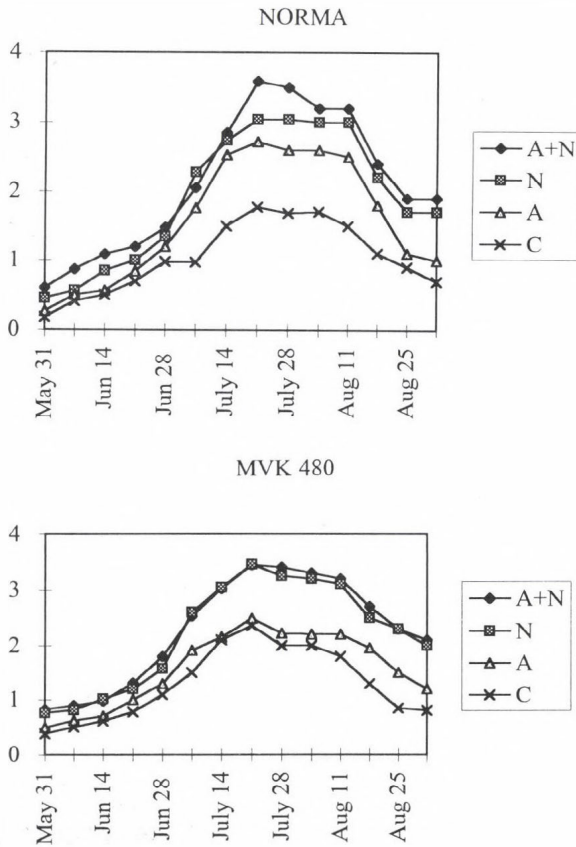


Fig. 1a. Seasonal variation in leaf area index of maize during 1996.

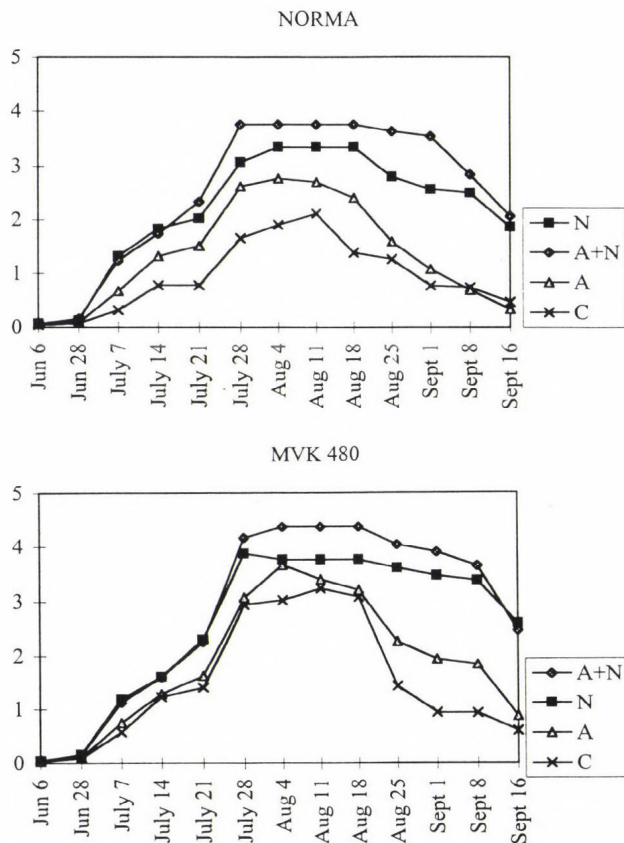


Fig. 1b. Seasonal variation in leaf area index of maize during 1997.

3.3 Evapotranspiration

Accumulated evapotranspiration from emergence until maturity is presented in Table 3. In both N fertilisation cases evapotranspiration is greater than the value of control case by 85–115 mm. Evapotranspiration was less when alginite was applied. Differences were 6.5–5.8% ($P < 0.01$) with N and 5.7–4.0% ($P < 0.05$) without N. In the case of cv. Norma differences were not significant. It suggests that the two hybrids response differently to alginite. While leaf area of cv. Norma became larger due to alginite, there was no significant increase in evapotranspiration. This indicated that alginite decreased transpiration rate in cv. Norma. Evapotranspiration water demand of MVK-480 was 6.4–8.1% ($P < 0.05$) higher than for cv. Norma in treatment C.

Table 3. Means of accumulated evapotranspiration (ET, mm) in 1996–97 for treatments and cultivars

Treatments	Cultivars		
	MVK-480	Norma	LSD _{0.05}
A+N	428.6	434.4	72.0 NS
N	450.7	439.6	4.3 *
LSD _{0.05}	17.0 *	59.3 NS	
A	337.4	343.1	4.1 *
C	359.5	335.1	9.2 *
LSD _{0.05}	7.1 *	12.3 NS	

* — significant at the 0.05 probability level; NS — non significant

Analyzing the evapotranspiration on daily basis showed that differences between treatments altered in time with and without alginite. Especially when the evaporative demand of the air was high, treatments with alginite had significant less evaporation, as it occurred during the first half of June in 1996 (Fig. 2). On 21 June 1996 a rainy period started and the differences in daily evapotranspiration between treatments were not so clear.

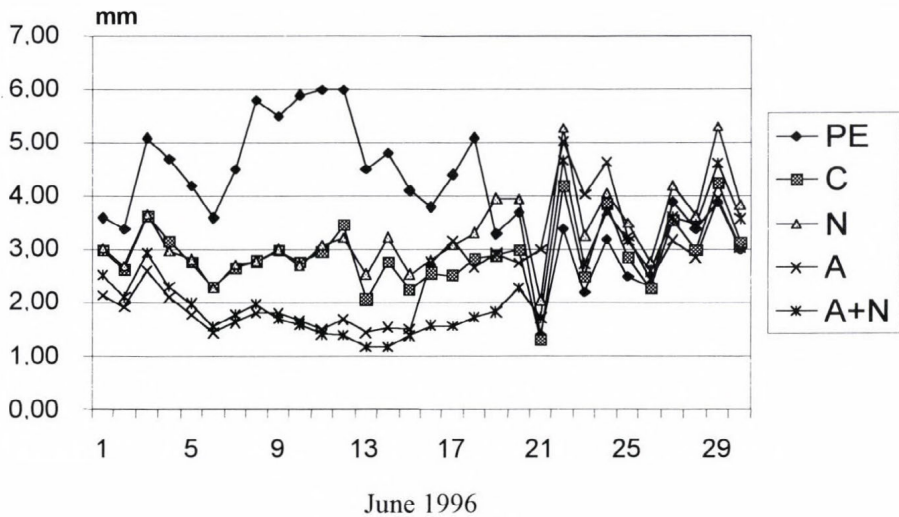


Fig. 2. Potential and actual daily evapotranspirations (mm) of cv. MVK-480 in different treatments (June 1996, Keszthely, Hungary).

3.4 Stomatal resistance

Stomatal resistance was measured at different phenological stages in 1997. In the beginning and the end of the season the daily means of stomatal resistance were about two times higher than in the middle of the vegetation period (Fig. 3). Minimum values were measured in the first half of August just after tasseling. At that time in daily course the values in the morning were between 3 and 4 s cm⁻¹, which fell down to 0.8–1.1 s cm⁻¹ around solar noon and then increased again up to 2–3 s cm⁻¹ in the afternoon.

In 1997 seasonal means of stomatal resistance of MVK-480 were 11.9–17.7% ($P < 0.05$), lower than averages of Norma in different treatments. Highest deviation was measured between control treatments. When N was also supplied, stomatal resistance values were closer to each other.

Significant increase in stomatal resistance between treatments A and without A was measured in the beginning of the growing season in accordance with decreasing water loss at that time. (This change ceased in seasonal average resistance of Norma but manifested in MVK-480.)

The nitrogen decreased the stomatal resistance in each treatment. Seasonal average resistance of Norma and MVK-480 with N were 24.6 and 20.5% ($P < 0.05$), less than in treatment C. When alginite and nitrogen were applied together, the size of change in mean stomatal resistance moderated in 14.8 and 9.0% (not significant) in Norma and MVK-480, respectively.

3.5 Microenvironment

Microenvironment in our terms means the characteristics of soil and air very close to the plants. We do not use the technical term of microclimate because of the small size of our experimental plots for developing a microclimate which differs from the macroclimate needs large homogeneous crop stand. Nevertheless any kind of physical or chemical interference may cause change in energy exchange inside plant canopy, which might be observed by measuring surface and air temperatures and humidity very close to the heat and water sources. Energy exchange is controlled mainly by radiation balance which is depending on the leaf area index. Therefore showing any other effect than the size of LAI is possible only when LAI is more or less the same. In our experiment it was the situation in cv. MVK-480, in both of the years in the treatments N and A+N. Therefore the alteration in microenvironment as a result of alginite application was more expressed in MVK-480, than those in variety of Norma. Daily course of the characteristics of the microenvironment is presented by values measured on 15 July of 1996. (Meteorological data for the sample day were as follows: 30.6°C maximum temperature; 13.6°C minimum temperature; 27.78 MJ m⁻² global radiation; 5.0 mm potential evapotranspiration. LAI in the treatments were as follows: A+N: 3.02; N: 3.04; A: 2.15; C: 2.1.) This day was a typical day considering analysis of the other days as well.

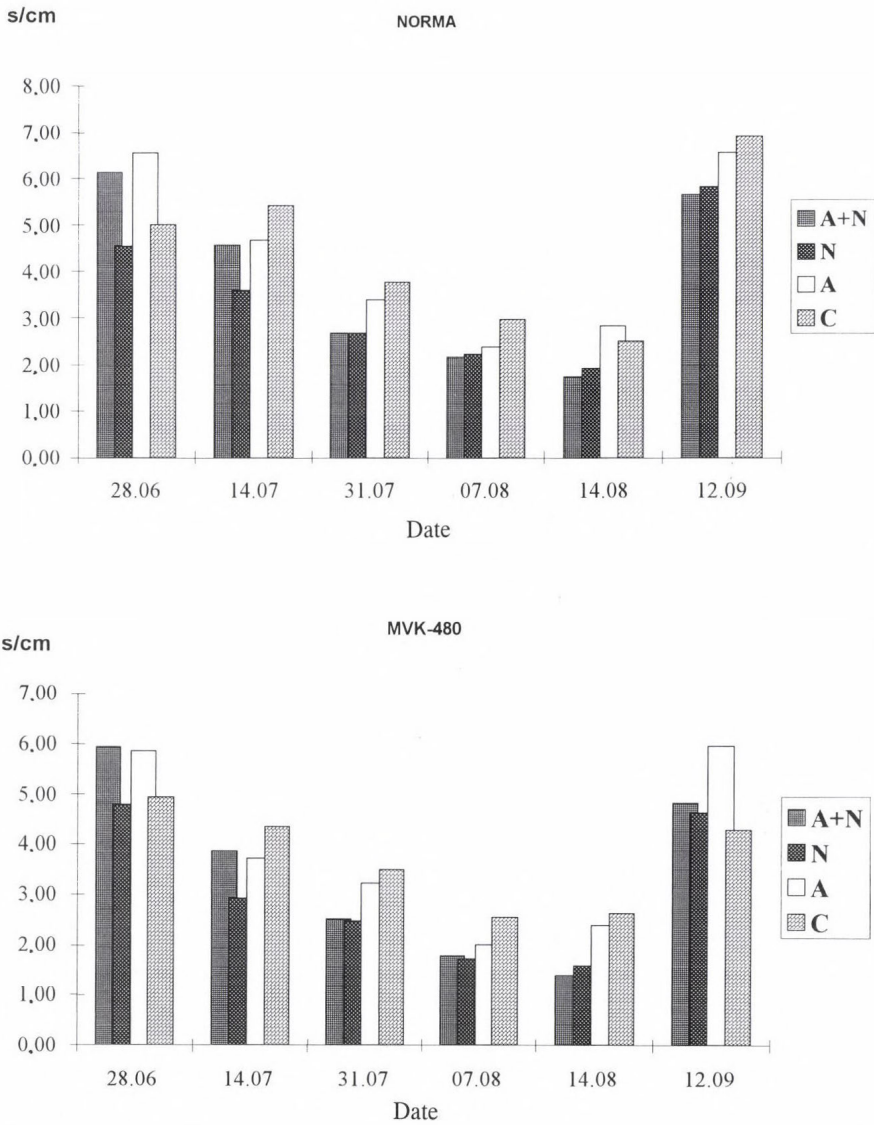


Fig. 3. Daily averages of stomatal resistance in Norma and in MVK-480 during 1997.

Soil surface temperatures did not differ between N and A+N. Differences were observed only between A and C, where the shadowing leaf area differed as well. Before 2 p.m. soil surface of A was cooler, but later in the afternoon A was the warmer treatment by some degrees (Fig. 4).

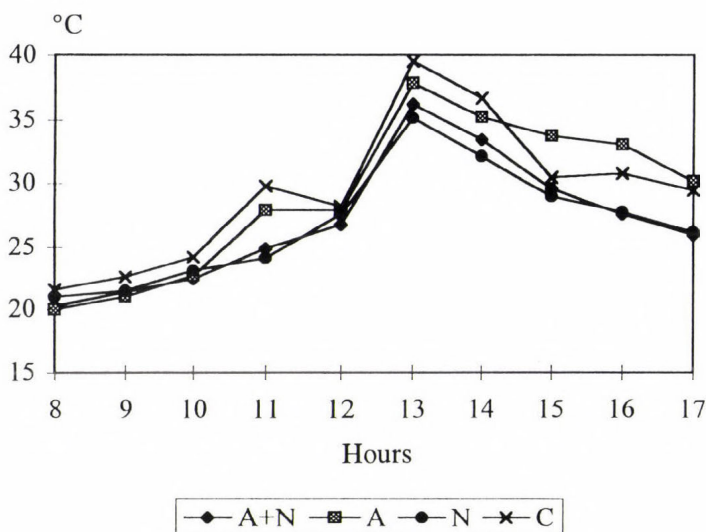


Fig. 4. Diurnal variation in soil surface temperature (15 July 1996).

The warmest air temperature was measured in the control treatment, independently on radiation angle of incidence (Fig. 5). In the morning (8.30–9.30 a.m.), closely to the soil surface the alginite decreased the air temperature in treatments A and A+N by 1.9°C and 0.6°C, compared to C and N, respectively. Later on, at higher solar angles treatment A caused the highest air temperature of all (4.6–5.1°C in different levels of the canopy), but the absolute value of its air temperature never exceeded the air temperature of control plots. It seems that both the warming and cooling processes derived from the soil are slower due to alginite application.

Air humidity was expressed by vapor pressure (hPa). The alginite increased the vapor pressure in each level of plant height (Fig. 6). The possible process which results this kind of profiles in temperature and humidity is the evapotranspiration, the sum of soil evaporation and plant transpiration. Parallel with growing LAI, the ratio of evaporation is decreasing. In spite of that we did not find significant differences in stomatal behaviour between the treatments — except of the beginning of the seasons —, the alginite decreases the transpiration of plants. This physiological influence might have been the reason of the changes of microenvironment conditions: higher air temperature is a result of the increased sensible heat flux derived from the surface to the ambient air. It is in accordance with a lower transpiration rate. Only the soil surface can provide excess humidity to the air within the canopy, because of the alginite content of the upper soil layer.

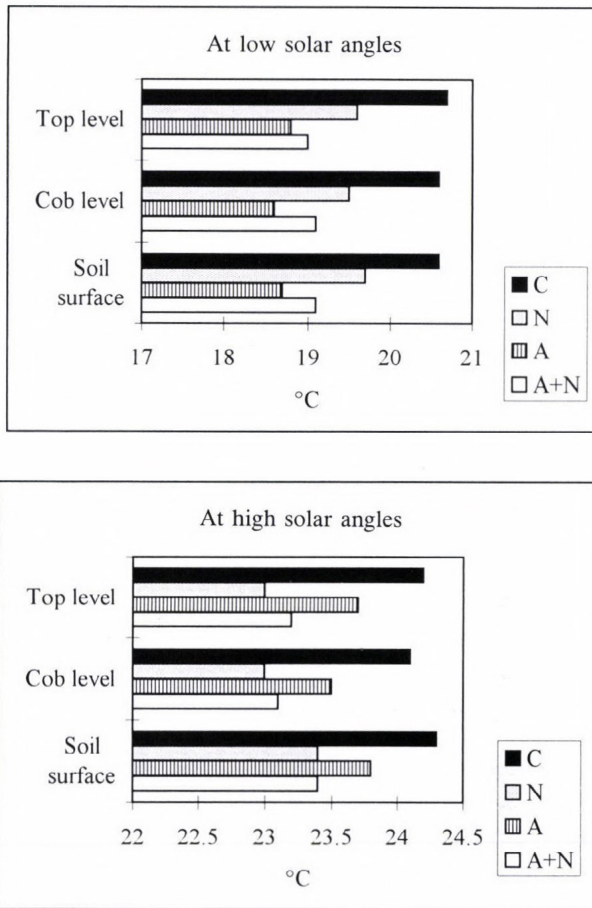


Fig. 5. Representative profiles of air temperature at low and high solar angles on the 15th July 1996.

3.6 Yield

MVK-480 produced more yield in both years and all treatments (*Table 4*). The differences compared to cv. Norma were between 23% and 75% depending on the treatment ($P < 0.05$). In the first year of the experiment there was no significant difference between the treatments N and A+N, but in the second year alginite increased cob yield by 13.4% ($P < 0.01$) and 36.7% ($P < 0.05$) in MVK-480 and Norma, respectively. The effect of alginite was more pronounced when no N was applied. In 1996 there was 7% ($P < 0.01$) and 10% ($P < 0.01$)

more yield in treatment A than in C for MVK-480 and Norma respectively. In 1997, A and C differed by 30% ($P < 0.05$) and 74% ($P < 0.05$), with yield levels that were less than in 1996. The higher differences in yield between A and C treatments in the second year might be caused by the gradual decreasing level of natural fertility of the soil because of no N application.

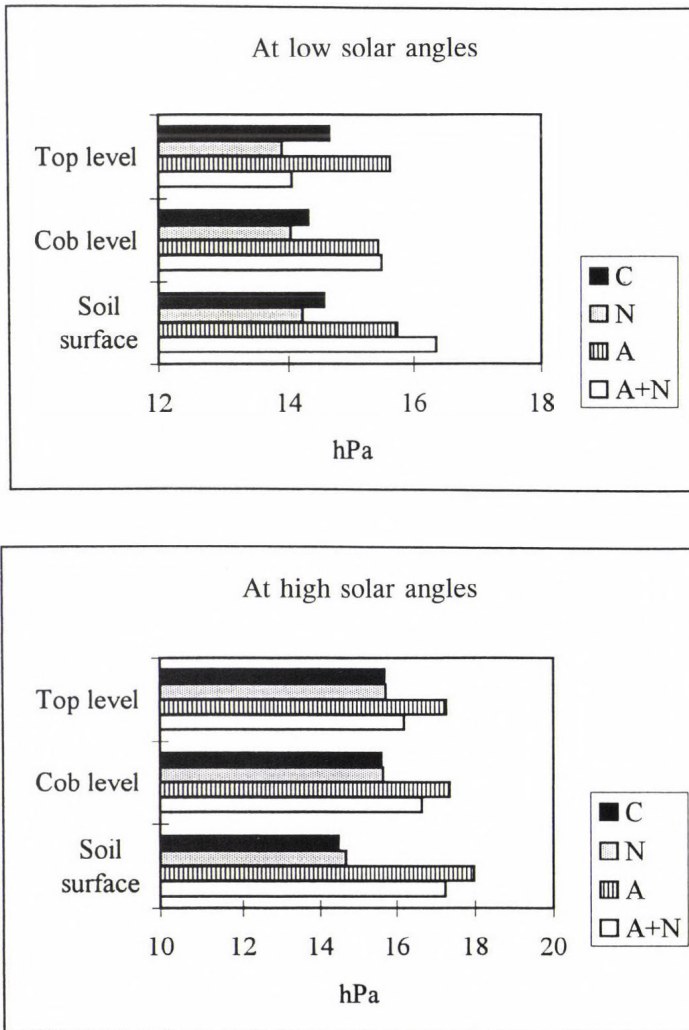


Fig. 6. Representative profiles of vapor pressure at low and high solar radiations on the 15th July 1996.

Table 4. Cob yield (g/m²) in ET pots for different hybrids in different treatments

Treatments	Cultivars		
	MVK-480	Norma	LSD _{0.05}
A+N	1271	998	352.0 NS
N	1194	845	365.5 NS
LSD _{0.05}	328.9 NS	316.1 NS	
A	566	378	485.9 NS
C	495	289	374.1 NS
LSD _{0.05}	94.6	206.4 NS	

4. Conclusions

In spite of high macroelement content of alginite, its available nutrient rate is relatively low. For this reason the alginite can not be taken into account as a complete nutrient source. The alginite is never suitable to substitute for fertilisers.

Considering the mixing ratio, the actual pH of the soil supplied with alginite has not been changed significantly.

The hybrids of maize MVK-480 and Norma are different in their water relations. Norma is taken to be a water stress tolerant variety while MVK-480 is sensitive to water shortage. Under well watered conditions the productivity of MVK-480 is greater than Norma as it was demonstrated by our experiments. The response of that two varieties to alginite treatment is also different. Alginite has a nutrient effect which is more effective in the case of Norma than in MVK-480, the increase in production and leaf area due to alginite is more expressive in Norma than in MVK-480. The effect of alginite on water relations manifested itself mainly in cv. MVK-480 where LAI did not change due to alginite while the evapotranspiration decreased.

Evapotranspiration consists of soil evaporation and plant transpiration. Stomatal resistance characterises the ability of plant for transpiring water through stomata. The actual evapotranspiration however depends on environmental factors as humidity and temperature of ambient air. Whilst we did not find unambiguous differences in stomatal behaviour resulted from alginite application between the individual treatments, the changes in air humidity as well as in soil and air temperatures indicate that physical conditions of evaporation has been modified because of alginite. Alginite is mixed in the upper soil level and makes changes in water capacity and conductivity through retaining water in this soil layer. This "wet sponge" provides a humid air inside the plant canopy which slows the soil heating processes. It has a feedback to the actual evapotranspiration especially when the potential evapotranspiration, calculated from climatological characteristics is extremely high.

The yield excess resulted from alginite supply is moderate. Considering the aim of the alginite application, use of environment friend agricultural technology on protected areas, alginite should therefore be regarded as a local mineral source of the Lake Balaton catchment area.

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NEWS

The WMO Norbert Gerbier – MUMM International Award winners in 1999

The fiftieth session of the WMO Executive Council conferred the Norbert Gerbier – MUMM International Award for 1999 on two Hungarian climate researchers, *Ms. Katalin Molnár* and *Mr. János Mika*. Both PhDs are presently affiliated at the Hungarian Meteorological Service.

The institution of the Norbert Gerbier – MUMM International Award was approved during the 39th session of the Executive Council, after the decease of Mr. Norbert Gerbier, the internationally appreciated French expert in agricultural meteorology. The purpose of the Award is to encourage and reward annually an original scientific paper on the influence of meteorology in a particular field of the physical, natural or human sciences, or conversely, the influence of one of these sciences on meteorology. The award aims at stimulating interest in such research, in support of WMO programmes.

Nomination for the Award is the right of all Permanent Representatives of Members of WMO, in response to the announcement annually distributed by the Secretary-General of the WMO, concerning papers published during the previous 18 months. The selection of the award winner is made by the selection committee, specially constituted for this purpose for a four-year period, that consists of not more than two members of the Executive Council and ex officio, the president of the Commission of Agricultural Meteorology. This decision is based on the preliminary assessment performed by three distinguished scientists in the field of meteorological applications.

The Award is presented annually and consists of a diploma, a medal bearing a likeness of Mr. Norbert Gerbier and a prize of 50.000 French francs.

The year 1999 Award was conferred on the authors for the paper entitled "Climate as a changing component of landscape: recent evidence and projections for Hungary" which was published in *Zeitschrift für Geomorphologie, N. F.*, Suppl.-Bd 110, (Berlin, Stuttgart), August, 1997.

The Award Ceremony took place during the fifty-first session of the Executive Council on 27 May 1999. Participants of the Award Ceremony were addressed by *Dr. J. Zillman*, President of the WMO; *Prof. G. O. P. Obasi*, Secretary-General of the WMO; *Mrs. G. Guiard-Gerbier*, France and *Mr. O. Brun*, representative of the MUMM Foundation, France. The Award was acknowledged by Katalin Molnár on behalf of the two winners who were accompanied by their spouses on this prominent occasion.

Summary of the
Norbert Gerbier – MUMM International Award '99
winner scientific paper

(The study was originally published in *Zeitschrift für Geomorphologie*
Neue Folge, Suppl.- Bd. 110, 1997, pp. 185-195, with 5 figures and 2 tables)

**Climate as a changing component of landscape:
recent evidence and projections for Hungary**

Katalin Molnár and János Mika

Summary. Recent and expected future changes of temperature (corrected for inhomogeneities) and precipitation in Hungary are investigated, also in connection with the global climate tendencies. These factors are generally assessed in long-term averages, an approach with conceptual difficulties if global climate really changes. Two periods (1900–1949 and 1950–1989) are identified to compare monthly temperature and precipitation averages for the two periods. Differences are considerable for both elements. In the second part of the paper, an attempt is made to describe the climatic conditions modified according to a presumed regional climate change scenario for the 230 micro-regions of Hungary. Mean temperatures for the summer and winter half-years as well as annual precipitation are first analysed for the period 1951–1980. Maps of future areal distribution of winter, summer and annual temperature and precipitation have been produced, assuming 0.5, 1, 2 and 4 K increase in the annual mean temperature of the Northern Hemisphere. Regional scenarios related to small (0.5 and 1 K) hemispherical changes, are based on the method of ‘slices’ which establishes bilinear regressions between local and hemispherical variations, after a special filtering. Consequences of larger global changes are derived from General Circulation Model (GCM) outputs and paleoclimatic analogues. An independent comparison of the 10 years (1981–1990) and 30 years (1951–1980, i.e., the reference period of the previous maps) is also presented to demonstrate that the regional climate of recent decade is not in contradiction with the scenarios fully based on data from the previous time periods. These maps also show that for studying geomorphologic processes characterised by time-scales not much longer than a decade, the precise derivation of climate averages is even more crucial.

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Volume 33 Number 5 1999

Fast-track

- D.J. Fish, D.E. Shallcross and R.L. Jones: The vertical distribution of NO₃ in the atmospheric boundary layer, 687-691.
- P. Wolkoff, P.A. Clausen, C.K. Wilkinson, K.S. Hougaard and G.D. Nielsen: Formation of strong airway irritants in a model mixture of (+)- α -pinene/ozone, 693-698.

Papers

- J. Kildesø, J. Vallarino, J.D. Spengler, H.S. Brightman and T. Schneider: Dust built-up on surfaces in the indoor environment, 699-707.
- M.W. Gardner and S.R. Dorling: Neutral network modelling and prediction of hourly NO_x and NO₂ concentrations in urban air in London, 709-719.
- E.D. Pellizzari, C.A. Clayton, C.E. Rodes, R.E. Mason, L.L. Piper, B. Fort, G. Pfeifer and D. Lynam: Particulate matter and manganese exposure in Toronto, Canada, 721-734.
- R.A. Hashmonay, M.G. Yost, Y. Mamane and Y. Benayahu: Emission rate apportionment from fugitive sources using open-path FTIR and mathematical inversion, 735-743.
- A.S. Ansarei and S.N. Pandis: Prediction of multicomponent inorganic atmospheric aerosol behavior, 745-757.
- E.J. Highwood, K.P. Shine, M.D. Hurley and T.J. Wallington: Estimation of direct radiative forcing due to non-methane hydrocarbons, 759-767.
- P.E. Padgett, E.B. Allen, A. Bytnerowicz and R.A. Minich: Changes in soil inorganic nitrogen as related to atmospheric nitrogenous pollutants in southern California, 769-781.
- M. Fang, M. Zheng, F. Wang, K.L. To, A.B. Jaafar and S.L. Tong: The solvent-extractable organic compounds in the Indonesia biomass burning aerosols — characterization studies, 783-795.
- W.O. Siegl, R.H. Hammerle, H.M. Herrmann, B.W. Wenclawiak and B. Luers-Jongen: Organic emission profile for a light-duty diesel vehicle, 797-805.
- J.D. Shannon: Regional trends in wet deposition on sulfate in the United States and SO₂ emissions from 1980 through 1995, 807-816.
- G.A. Allen, J. Lawrence and P. Koutrakis: Field validation of a semi-continuous method for aerosol black carbon (aethalometer) and temporal patterns of summertime hourly black carbon measurements in southwestern PA, 817-823.
- J.A. Lind and G.L. Kok: Emission strengths for primary pollutants as estimated from an aircraft study of Hong Kong air quality, 825-831.
- S.E. Conell and T.D. Jickells: Water-soluble organic nitrogen in atmospheric aerosol: a comparison of UV and persulfate oxidation methods, 833-840.

Volume 33 Number 6 1999

- H. Zhuang, C.K. Chan, M. Fang and A.S. Wexler:* Size distributions of particulate sulfate, nitrate, and ammonium at a coastal site in Hong Kong, 843-853.
- J.P. Greenberg, A. Guenther, P. Zimmermann, W. Baugh, C. Geron, K. Davis, D. Helmig and L.F. Klinger:* Tethered balloon measurements of biogenic VOCs in the atmospheric boundary layer, 855-867.
- M.D. Moulik and J.B. Milford:* Factors influencing ozone chemistry in subsonic aircraft plumes, 869-880.
- T.C.W. Tung, C.Y.H. Chao and J. Burnett:* A methodology to investigate the particulate penetration coefficient through building shell, 881-893.
- A. Kasper-Giebl, M.F. Kalina and H. Puxbaum:* Scavenging ratios for sulfate, ammonium and nitrate determined at Mt. Sonnblick (3016 m a.s.l.), 895-906.
- L.W.A. v. Hove, M.E. Bossen, F.A.M. de Bok and C.A.M. Hooijmaijers:* The uptake of O₃ by poplar leaves: the impact of a long-term exposure to low O₃-concentrations, 907-917.
- E.G. Pavelin, C.E. Johnson, S. Rughooputh and R. Toumi:* Evaluation of pre-industrial surface ozone measurements made using Schönbein's method, 919-929.
- P.A. Ariya, H. Niki, G.W. Harris, K.G. Anlauf and D.E.J. Worthy:* Polar sunrise experiment 1995: hydrocarbon measurements and tropospheric Cl and Br-atoms chemistry, 931-938.
- I. Mavroidis, R.F. Griffiths, C.D. Jones and C.A. Biltoft:* Experimental investigation of the residence of contaminants in the wake of an obstacle under different stability conditions, 939-949.
- S. Parat, A. Perdrix, S. Mann and P. Bacconnier:* Contribution of particle counting in assessment of exposure to airborne microorganisms, 951-959.
- J.S. Fuglestad, T.K. Bernsten, I.S.A. Isaksen, H. Mao, X.-Z. Liang and W.-C. Wang:* Climatic forcing of nitrogen oxides through changes in tropospheric ozone and methane; global 3D model studies, 961-977.
- E. Dupont, L. Menut, B. Carissimo, J. Pelon and P. Flamant:* Comparison between the atmospheric boundary layer in Paris and its rural suburbs during the ACLAP experiment, 979-994.
- U. Kuhm, C. Amman, A. Wolf, F.X. Meixner, M.O. Andreae and J. Kesselmeier:* Carbonic sulfide exchange on an ecosystem scale: soil represents a dominant sink for atmospheric COS, 995-1008.

Technical Note

- X. Li-Jones, C. McCormick and H.B. Maring:* Zero offset in computer-recorded analog data in measuring aerosol scattering coefficient: a "threshold" or a "blank"? 1009-1013.

Volume 33 Number 7 1999

- A.T. Buckland and D.R. Middleton:* Nomograms for calculating pollution within street canyons, 1017-1036.
- R.M. Harrison, J.P. Shi and M.R. Jones:* Continuous measurements of aerosol physical properties in the urban atmosphere, 1037-1047.
- J.G. Vasilou, D. Sorensen and P.H. McMurry:* Sampling at controlled relative humidity with a cascade impactor, 1049-1056.
- R.D. Saylor, W.I. Chameides and M.E. Chang:* Demonstrating attainment in Atlanta using urban airshed model simulations: impact of boundary conditions and alternative forms of the NAAQS, 1057-1064.
- B. Eklund:* Comparison of line- and point-source releases of tracer gases, 1065-1071.

- D.J. Luecken, G.S. Tonnesen and J.E. Sickles, II:* Differences in NO_y speciation predicted by three photochemical mechanisms, 1073-1084.
- M. Kahn, Y.J. Yang and A.G. Russell:* Photochemical reactivities of common solvents: comparison between urban and regional domains, 1085-1092.
- A. Venkatram, D. Fitz, K. Bumiler, S. Du, M. Boeck and C. Ganguly:* Using a dispersion model to estimate emission rates of particulate matter from paved roads, 1093-1102.
- C. Treviño and F. Méndez:* Simplified model for the prediction of ozone generation in polluted urban areas with continuous precursor species emissions, 1103-1110.
- Y. Andersson-Sköld and D. Simpson:* Comparison of the chemical schemes of the EMEP MSC-W and IVL photochemical trajectory models, 1111-1129.
- C. Zellweger, M. Ammann, P. Hofer and U. Baltensperger:* NO_y speciation with a combined wet effluent diffusion denuder-aerosol collector coupled to ion chromatography, 1131-1140.
- I.J. Simpson, G.C. Edwards and G.W. Thurtell:* Variations in methane and nitrous oxide mixing ratios at the Southern boundary of a Canadian boreal forest, 1151-1158.
- M.A.K. Khalil and R.A. Rasmussen:* Atmospheric chloroform, 1141-1158.
- C.M. Mitic, W.J. Massman, P.H. Schuepp and J.L. Collett Jr.:* Structural analysis and flux associations of CO₂, H₂O, heat and ozone over cotton and grape canopies, 1159-1173.

Volume 33 Number 8 1999

- M. Glasius, M.F. Carlsen, T.S. Hansen and C. Lohse:* Measurements of nitrogen dioxide on Funen using diffusion tubes, 1177-1185.
- D.J. Hall, S. Walker and A.M. Spanton:* Dispersion from courtyards and other enclosed spaces, 1187-1203.
- Z. Guo, J.C.S. Chang, L.E. Sparks and R.C. Fortmann:* Estimation of the rate of VOC emissions from solvent-based indoor-coating materials based on product formulation, 1205-1215.
- H.N. Knudsen, U.D. Kjaer, P.A. Nielsen and P. Wolkoff:* Sensory and chemical characterization of VOC emissions from building products: impact of concentration and velocity, 1217-1230.
- A. Feilberg, R.M. Kamens, M.R. Strommen and T. Nielsen:* Modeling the formation, decay, and partitioning of semivolatile nitro-polycyclic aromatic hydrocarbons (nitronaphthalenes) in the atmosphere, 1231-1243.
- J.M.F. Diaz, M.A.R. Braña, B.A. Garcia, C.G.-P. Muñoz and P.J.G. Nieto:* Difficulties inherent to the use of analytic solution of the condensation-evaporation equation for multicomponent aerosols, 1245-1259.
- L. Morawska, S. Thomas, D. Gilbert, C. Greenaway and E. Rijnders:* A study of the horizontal and vertical profile of submicrometer particles in relation to a busy road, 1261-1274.
- M.E. Jenkin and G.D. Hayman:* Photochemical ozone creation potentials for oxygenated volatile organic compounds: sensitivity to variations in kinetic and mechanistic parameters, 1275-1293.
- E. Smet, H.V. Langenhove and I.D. Bo:* The emission of volatile compounds during the aerobic and the combined anaerobic/aerobic composting of biowaste, 1295-1303.
- M.A.K. Khalil and R.A. Rasmussen:* Atmospheric methyl chloride, 1305-1321.

Technical Note

- A. Kasper-Giebl and H. Puxbaum:* Deposition of particulate matter in diffusion tube samplers for the determination of NO₂ and SO₂, 1323-1326.
- M. Sharan, A.K. Yadav, M.P. Singh and S. Gupta:* Accounting for the source strength in the solution of the diffusion equation: Alternative mathematical formulations, 1327-1330.

Volume 33 Number 9 1999

- R. von Glasow and A. Bott*: Interaction of radiation fog with tall vegetation, 1333-1346.
- R. Gabriel, L. Schäfer, C. Gerlach, T. Rausch and J. Kesselmeier*: Factors controlling the emissions of volatile organic acids from leaves of *Quercus ilex* L. (Holm oak), 1347-1355.
- R. Leuning, S.K. Baker, I.M. Jamine, C.H. Hsu, L. Klein, O.T. Denmead and D.W.T. Griffith*: Methane emission from free-ranging sheep: a comparison of two measurement methods, 1357-1365.
- H.V. Andersen, M.F. Hovmand, P. Hummelshøj and N.O. Jensen*: Measurements of ammonia concentrations, fluxes and dry deposition velocities to a spruce forest 1991-1995, 1367-1383.
- M. Väkevä, K. Hämeri, M. Kulmala, R. Lahdes, J. Ruuskanen and T. Laitinen*: Street level versus rooftop concentrations of submicron aerosol particles and gaseous pollutants in an urban street canyon, 1385-1397.
- G. Åberg, D. Stijfhoorn, K. Iden and R. Löfvendahl*: Carbon isotope exchange during calcite sulphation, 1399-1402.
- A.N. Thakur, H.B. Singh, P. Mariani, Y. Chen, Y. Wang, D.J. Jacob, G. Brasseur, J.-F. Müller and M. Lawrence*: Distribution of reactive nitrogen species in the remote free troposphere: data and model comparisons, 1403-1422.
- A. Calogirou, B.R. Larsen and D. Kotzias*: Gas-phase terpene oxidation products: a review, 1423-1439.
- K.A. Rahn*: A graphical technique for determining major components in a mixed aerosol. I. Descriptive aspects, 1441-1455.
- M. Ikegawa, M. Kimura, K. Honda, I. Akabane, K. Makita, H. Motoyama, Y. Fujii and Y. Itokawa*: Geographical variations of major and trace elements in East Antarctica, 1457-1467.
- Y. Deng and Y. Zuo*: Factors affecting the levels of hydrogen peroxide in rainwater, 1469-1478.
- F. Sunny, T.N. Mahadevan and V. Sitaraman*: Estimation of actinic flux and photolysis rate constant of NO_2 from aerosol size data, 1479-1488.

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