

# IDŐJÁRÁS

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# IDŐJÁRÁS

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# IDŐJÁRÁS

*Quarterly Journal of the Hungarian Meteorological Service*  
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## **A preliminary estimation of the nitrogen compounds exchange between the atmosphere and a Spruce forest**

**László Horváth<sup>1</sup>, Soren Christensen<sup>2</sup>, Ernő Führer<sup>3</sup>, Sugárka Kelecsényi<sup>4</sup>,  
Róbert Mészáros<sup>5</sup>, Zoltán Nagy<sup>1</sup> and Tamás Weidinger<sup>5</sup>**

<sup>1</sup>*Hungarian Meteorological Service,*

*H-1675 Budapest, P.O. Box 39, Hungary; e-mail: lhorvath@met.hu*

<sup>2</sup>*University of Copenhagen, Zoological Institute, Department of Population Biology,  
15 Universitetsparken, 2100 Copenhagen, Denmark*

<sup>3</sup>*Forest Research Institute, 1023 Budapest, Frankel Leó u. 42/44, Hungary*

<sup>4</sup>*Ministry for Environmental Protection, 1117 Budapest, Fő u. 44/50, Hungary*

<sup>5</sup>*Eötvös Loránd University, Department of Meteorology,  
1117 Budapest, Pázmány Péter sétány 1, Hungary*

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**Abstract**—On the basis of a one and a half year investigation carried out at a forestry station in Hungary in a Norway Spruce stand during 1996–97 the net dry deposition (the difference of the dry deposition and the emission) is  $1.8 \text{ gN m}^{-2} \text{ yr}^{-1}$ . As a consequence of the nitrous oxide and nitric oxide emission from the forest soil only a few percent of the deposited nitrogen compounds is released back to the atmosphere. The wet deposition amounts to approximately one half of the dry deposition,  $0.9 \text{ gN m}^{-2} \text{ yr}^{-1}$ . The total dry+wet deposition is  $2.7 \text{ gN m}^{-2} \text{ yr}^{-1}$ . From the difference between the measured dry deposition and the throughfall + stemflow measurements, it seems that one half of the deposited nitrogen compounds is taken up by stomata, the other half is leached to the forest soil.

**Key-words:** nitrogen balance, atmospheric wet and dry deposition, forest ecology, throughfall deposition.

### ***1. Introduction***

The role of atmospheric pollutants in forest health is well known. Acid deposition, especially near the densely populated regions of North-America and Europe, is accompanied by a certain amount of forest damage. The supply of nitrogen, phosphorus and other nutrients to the forest ecosystem mainly takes place from the atmosphere resulting in serious nutrient overloading at some places on the Earth (e.g., in The Netherlands).

During the intensive exchange processes of the trace materials between the atmosphere and the forest ecosystems, the forest (including the forest soil) may be a source of a certain group of compounds (e.g., nitric oxide, nitrous oxide, isoprene, terpenes). On the other hand, in the case of some materials, the atmosphere can also be an important source for the forest. Some of them provide nutrients (especially nitrogen compounds) to the plants, others are harmful to the vegetation (e.g., oxidants and acidic compounds).

The forest ecosystem may act as a source or a sink for nitrogen compounds as a function of the physical and chemical circumstances. Nitrogen compounds, e.g. nitric oxide, nitrogen dioxide, ammonia, nitric acid and particulate nitrates and ammonium also take part in the acidification and nutrient supply or loading. Through the decomposition of the organic matter content in the forest soil, as a result of denitrification processes, nitrous oxide is produced and emitted. The rate of nitrous oxide emission is proportional to the temperature, humidity and organic matter content of the soil (*Christensen et al.*, 1996). In certain circumstances the forest soil emits a substantially high amount of nitrous oxide, which may partly counterbalance the deposition of the other nitrogen compounds.

Nitrogen compounds, similarly to other pollutants, are deposited by turbulent diffusion motions followed by quasi-laminar, molecular diffusion. Reaching the surface of the vegetation, they are absorbed on the leaves or are taken up by the stomata. A limited amount of adsorption on the trunk and the soil surface is also possible. The materials taken up by cuticular adsorption are leached by the following precipitation event and appear in the throughfall precipitation samples. The results concerning the dry deposition of reactive nitrogen compounds in forests are compiled by *Hansson and Lindberg* (1991).

Nitrogen dioxide can be emitted from the surface of the canopy in special circumstances (*Bowden*, 1986). The nitric oxide emitted from the forest soil by bacterial activity is transformed by the chemical reaction with ozone into nitrogen dioxide that may modify the vertical profiles of nitrogen oxides in and above the canopy (*Duyzer*, 1991). Because the intensity of the solar radiation is relatively low below the canopy, the decomposition of nitrogen dioxide by photolysis into nitric oxide is limited there. The nitrogen dioxide produced by the reaction of ozone and nitric oxide is partly taken up by the canopy. If the concentration of nitrogen dioxide is higher below the canopy than above, upward flux is probable, otherwise downward flux, i.e. deposition can be detected. Because the two processes, namely the upward and downward flux, are in competition, the so-called net flux may be bi-directional above forests, i.e. both net deposition and emission can be observed. For example, nitrogen oxides are mostly deposited especially at high atmospheric concentrations and during intensive uptake of nitrogen by the vegetation.

As for the other important group, the reduced nitrogen compounds (ammonium and ammonia) can partly be emitted or deposited. Ammonia, produced by denitrification processes in the forest soil, can be emitted. The

ammonia gas emitted from the soil is quickly taken up in the canopy. There is a so-called compensation point concentration of ammonia controlled by and in equilibrium with the canopy (Farquhar *et al.*, 1980). When the atmospheric ammonia level is higher than the compensation point, net deposition and in the opposite case emission is expected. According to recent measurements the rate of ammonia uptake is more substantial in the forest as was previously expected (Duyzer *et al.*, 1994; Wyers *et al.*, 1993).

It follows that the estimation of the nitrogen balance between the forest and the atmosphere is rather complicated. It requires the determination of the fluxes of both oxidised and reduced gas phase nitrogen compounds and particulate nitrate and ammonium. Beside that, the determination of the rate of the wet deposition of nitrogen compounds is also necessary. For the deposition estimation wet deposition, throughfall and stemflow measurements are also required. The measurement of the emissions of nitrogen compounds from the forest soil is also important.

A joint research program was started by the Forest Research Institute and the Hungarian Meteorological Service in 1988 to determine the rate of the acid deposition in forests. The investigations concentrated on nitrogen and sulphur compounds (Führer and Horváth, 1990, 1992; Führer *et al.*, 1994; Horváth *et al.*, 1993a, 1994a; Horváth and Führer, 1991). In these measurements the concentrations of the most important nitrogen and sulphur compounds and the rate of wet and dry deposition were determined. Dry deposition was estimated by using a simple inferential model taking into consideration average dry deposition velocity figures from the literature determined for surfaces different from the forest. This approximation involves some uncertainty in the estimation of dry deposition fluxes. Many results suggest that the dry deposition velocity of nitrogen compounds to the forest is substantially higher than was previously estimated, especially for nitrate and ammonium particles. On the other hand, in some cases (e.g., for nitrogen oxides) emission is also to be expected, which was not taken into account in our calculations before either. Therefore, the re-evaluation of the rate of dry+wet deposition of nitrogen compounds is necessary. In the frame of an international project (EUREKA, EUROTRAC, BIATEX) the dry deposition velocities of the ozone, nitrogen oxides and sulphur dioxide were determined in a Norway Spruce forest by the gradient method during field campaigns carried out in the four different seasons (Horváth, 1992, 1993; Horváth *et al.*, 1992, 1993b,c, 1994b,c,d; Weidinger and Horváth, 1994). It was found that the dry deposition velocity of nitrogen oxides strongly depends on meteorological circumstances. In some cases e.g., in the summer of 1993 and 1994 more emission than deposition was observed (Horváth *et al.*, 1995a,b, 1997).

For these reasons our hypothesis is that the nitrogen balance may be different from the figures that were previously determined. From the point of view of the acidification or for the determination of the nutrient loading, we

need to know the nitrogen balance between the forest and the atmosphere more accurately.

The aim of our investigation is to determine the nitrogen balance between the atmosphere and the forest taking into account all nitrogen compounds existing both in reduced and oxidised forms in the gaseous or aerosol phase. The estimation of the nitrogen balance may also allow us to decide whether the deposited nitrogen compounds are being accumulated in the forest soil or that the emission processes are decreasing the effect of deposition partly or totally.

## 2. Measurements

For the estimation of the nitrogen balance between the atmosphere and the forest examined, we have to determine the dry and wet deposition rates of the different nitrogen compounds. In the case of the compounds being deposited or emitted the measurement of the rate of net flux is also necessary. The determination of the dependence of the rate of deposition on the meteorological conditions is also required.

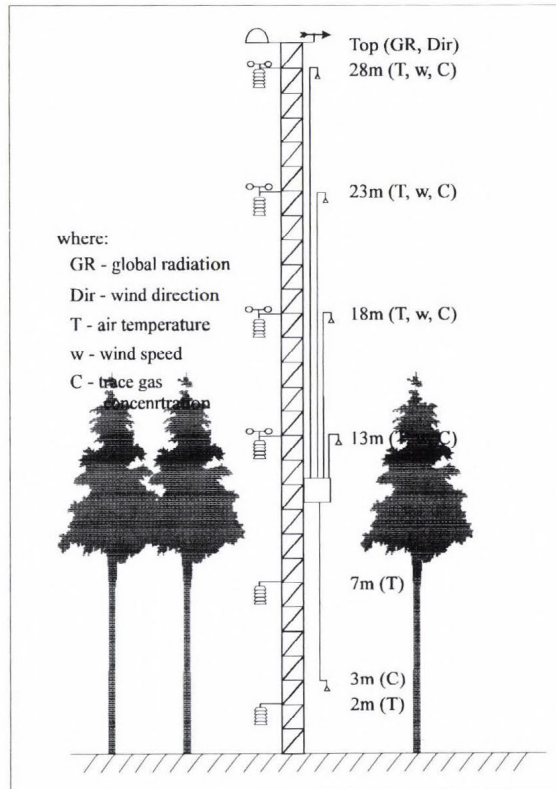
Parallel with the determination of the dry flux of the nitrogen compounds, the difference between wet deposition and throughfall + stemflow measurements also has to be taken into consideration.

Flux measurements were carried out at the Forest Research Institute's measuring site in the Mátra Mountains (Nyírjes station,  $\varphi = 47^{\circ}54'N$ ;  $\lambda = 19^{\circ}57'E$ ;  $h = 600$  m a.s.l.) in a Norway Spruce forest. The area of the forest is approximately 50 ha, the mean altitude is 600 m. The surrounding forests consist of mostly Pine and Beech species. The average height of the stand is about 16 meters, with a leaf area index of 3.3. Concentration measurements were carried out near the forest at an open place. During the year of 1996 the concentration of nitrogen compounds (ammonia, nitric acid, nitric oxide, nitrogen dioxide, ammonium and nitrate particles) as well as the ammonium and nitrate content of the precipitation water was measured for the determination of the wet deposition.

Gas and aerosol sampling was carried out by a three stage filter pack on the basis of 24 hour continuous sampling. The first Teflon filter captures the aerosol particles, the second and third Whatman filters prepared by basic and acidic materials collect the acidic (nitric acid) and alkaline (ammonia) gases, respectively. The sampling and analytical procedures were performed by the standard method proposed by the *EMEP* (1996) sampling and analysis protocol. The sampling of the precipitation water was carried out by a wet only precipitation sampler.

Concentrations of nitrogen compounds were determined as ammonium and nitrate ions by spectrophotometric and ion chromatographic methods, respectively. In the year of 1996 the concentration profile and the flux of ozone, nitric

oxide and nitrogen dioxide below, in and above the canopy were continuously monitored. The flux measurements were carried out by the gradient method described in *Horváth et al. (1998)*, by the tower shown on *Fig. 1*.



*Fig. 1.* The scheme of the measuring tower at Nyírjes station.

The gradient method is based on the parallel determination of the eddy diffusivity ( $K$ ) and the vertical profile (gradient) of the concentration on the basis of the following expression:

$$F = -K \frac{\Delta C}{\Delta z}, \quad (1)$$

where the second term on the right denotes the gradient of the concentration.

The eddy diffusivity was calculated from the measured wind and temperature profiles on the basis of Monin-Obukhov's semi-empirical similarity theory (for details refer to *Horváth et al.*, 1998). The rate of eddy diffusivity depends on the roughness of the surface and the meteorological conditions, especially the stratification. In stable cases (mostly during the night-time hours), for energy reasons, the rate of exchange is low, when the eddy diffusivity is generally lower by one order of magnitude comparing with the unstable cases (daytime hours).

The concentration gradients were determined by sequential measurements at five different levels on the basis of five minute averaging (see on Fig. 1). The air was passed from the different levels to the HORIBA NO/NO<sub>x</sub> gas monitor. The monitor was calibrated every two weeks.

The determination of nitrous oxide flux from the forest soil surface was carried out by the chamber method with a 10 cm diameter frame pressed 5 cm deep into the forest floor closed with a lid sealing 0.5 l of air during the measurement. In the chamber, installed on the forest soil, the accumulation of nitrous oxide was measured at 0, 15 and 30 minutes after closing. Samples were taken by evacuated tubes and were analysed by gas chromatography. The emission rate from the soil was calculated from the accumulation rate of the nitrous oxide in the chamber (*Ambus and Christensen*, 1995). Twenty four chambers were installed in the forest near the measuring tower in a net of 10 × 10 metres. Measurements were performed at three occasions on 15–16 May 1997. Simultaneously with the chamber measurements, five or ten replicate air samples were collected at the four heights from the tower immediately prior to and following the chamber measurements. Due to the low precision of the gas chromatograph when measuring atmospheric nitrous oxide gradients that require accuracy in the ppb range only data from the last of the three samplings with three replicate concentration measurements at each elevation will be reported. Other complementary measurements like wind direction, air humidity (at three levels), radiation balance and global radiation, soil moisture and humidity were also completed.

The data from sensors and the gas monitor was treated by a computer equipped data acquisition system. Five minute averages of the data have been stored.

### ***3. The nitrogen balance***

#### *3.1 The flux of nitric oxide and nitrogen dioxide*

On the basis of the concentration, wind and temperature profile measurements carried out during the year of 1996, the flux of nitric oxide and nitrogen dioxide was determined as described in Section 2, above the Norway Spruce forest, in the layer between 18 and 23 metres. This is the layer where the effect

of deposition or emission processes can be detected. The main results are summarized in *Table 1*, where the average figures for the different quarters of the year can be seen separately for stable (positive Richardson number) and for unstable (negative Richardson number) stratification cases. During daytime hours when the energy balance for the surface is positive the stratification is generally unstable, while in night hours stable stratifications are expected. In the daytime, when the turbulent exchange processes are generally more intensive, a higher exchange rate (emission or deposition) can be observed in the table. The emission cases are marked by a negative sign. Because the emission and deposition processes take place in parallel for both nitric oxide and nitrogen dioxide, the so-called net flux can be detected during the measurements. In stable cases emission only is observed for nitric oxide (*Table 1*, 2nd column). This may be the consequence of the forest soil emission. Moreover, in stable stratification cases the stomata are closed therefore the NO uptake (deposition) is limited. For NO the only deposition form possible is stomatal uptake as it is practically insoluble in water, and thus the irreversible adsorption on the wet surface of the leaves is limited. In daytime hours, as a consequence of the photosynthetic active radiation, when the stratification is generally unstable, the deposition and emission processes are in competition, therefore both deposition and emission can be measured (*Table 1*, 3rd column).

*Table 1.* Flux of nitrogen oxides at Nyírjes monitoring station in  $\text{ng m}^{-2} \text{s}^{-1}$

Period	NO flux		NO <sub>2</sub> flux	
	R <sub>i</sub> >0	R <sub>i</sub> <0	R <sub>i</sub> >0	R <sub>i</sub> <0
1st quarter	-2.1 (2 982)	-13.0 (771)	2.0 (3 239)	-21.7 (784)
2nd quarter	-1.4 (1 944)	-13.0 (845)	1.1 (2 763)	22.8 (1 336)
3rd quarter	-4.6 (1 172)	7.2 (734)	3.3 (2 204)	48.4 (1 162)
4th quarter	-6.0 (1 056)	12.4 (767)	14.4 (1 886)	52.3 (1 030)

R<sub>i</sub>>0 and R<sub>i</sub><0 indicates stable and unstable cases, respectively. In brackets the number of cases are indicated, negative and positive signs denote the emission and deposition cases, respectively.

In the case of nitrogen dioxide mostly deposition can be observed. This is obvious since the forest soil is not the source of nitrogen dioxide. In some cases, however, on the surface of leaves chemical reactions take place leading to the release of nitrogen dioxide (*Bowden, 1986*). The emission figure observed in the first quarter of the year may be attributed to this phenomenon.

The profiles of nitric oxide and nitrogen dioxide are controlled not only by emission and deposition processes, but also by the chemical reaction of nitric oxide with ozone producing nitrogen dioxide and by the photolysis of the nitrogen dioxide. This may lead to the overestimation of the nitric oxide flux and the underestimation of the nitrogen dioxide flux, however, the estimation of the total flux (sum of nitric oxide and nitrogen dioxide flux) is probably correct.

On a yearly average nitric oxide emission and nitrogen dioxide deposition can be detected at the station.

### 3.2 *The flux of ammonia/ammonium and nitric acid/nitrate*

Concentrations of ammonia gas, nitric acid vapor, as well as nitrate and ammonium particles were continuously measured on the basis of 24 hour sampling at Nyírjes station. In contrast with nitrogen oxides, direct flux measurements were not carried out from a practical point of view. In the future by improving the measuring system the direct determination of dry deposition velocities of these components will be also available.

In the case of ammonia/ammonium and nitric acid/nitrate the dry flux can be inferred by using the concentrations if the average dry deposition velocities representative of the examined surface are known. The flux thus can be estimated by the  $F = -v_d C$  expression, where  $v_d$  is the dry deposition velocity,  $C$  is the measured concentration.

The dry deposition velocity of ammonia gas to the spruce forest depends on the season. Because the ammonia is mostly taken up through stomata, the rate of the flux depends on the season i.e. on the open or closed state of the stomata. According to measurements above pine forest (*Andersen et al.*, 1993) the dry deposition velocity of ammonia ranges between  $-0.125$  and  $0.201 \text{ m s}^{-1}$ , with an average of  $0.026 \text{ m s}^{-1}$ .

In the vegetation period when nitrogen uptake is at the maximum the average ammonia flux is  $87 \mu\text{gN m}^{-2} \text{ h}^{-1}$ , which refers to a  $0.045 \text{ m s}^{-1}$  deposition velocity at average concentration. The result of another investigation shows that the ammonia dry deposition velocity is in the range of  $0.02\text{--}0.03 \text{ m s}^{-1}$  at a yearly average in a spruce forest (*Duyzer et al.*, 1994).

The dry deposition velocity figures of the nitric acid determined for pine forests are extremely high,  $0.07 \text{ m s}^{-1}$  (*Janson and Granat*, 1997). There is no expressed difference between the daytime and night-time figures, which suggests that cuticular adsorption is the dominant deposition process in comparison with stomatal uptake. Probably there is no large difference between summer and winter figures in pine forests.

The aerosol particles are generally captured by the surface of leaves (cuticle) while the stomatal uptake has lower importance. Therefore the seasonal variation of the deposition velocity is not so expressed. Previously, the dry

deposition velocity of particles was determined by theoretical calculations and by wind tunnel experiments, resulting in substantially lower deposition figures than were recently determined by experiments. In our previous calculations (Horváth *et al.*, 1993a) the lower figures were used in the calculation of the acid (sulphur and nitrogen) deposition at Nyírjes station. In recent years the contradiction has become obvious between the results of the two methods (Ruijgrok *et al.*, 1993; Borrell *et al.*, 1997). Different research groups agree that the figures determined experimentally are correct and they suggest  $>0.01 \text{ m s}^{-1}$  dry deposition velocity for forests in the case of particles.

The dry fluxes of ammonia, nitric acid and nitrate, ammonium particles were determined by the recently determined deposition figures using the average concentrations in *Table 2*. The calculated dry fluxes can be seen in *Table 3*.

*Table 2.* The concentration of nitrogen compounds at Nyírjes monitoring station in  $\mu\text{g m}^{-3}$

Period	Concentration					
	Nitric oxide	Nitrogen dioxide	Ammonia	Nitric acid	Ammonium	Nitrate
1st quarter	0.69	8.06	0.52*	1.51	0.42	0.54
2nd quarter	0.57	3.28	1.68*	0.50	0.56	0.50
3rd quarter	0.47	3.45	0.92*	1.50	0.70	0.59
4th quarter	0.62	3.98	0.41	1.87	0.86	1.39
1996	0.59	4.69	0.88	1.35	0.64	0.76

\*In the cases marked by an asterisk there were no ammonia measurements for technical reasons. Data replaced by the results of another forestry station in Hungary (K-pusztá).

### 3.3 The flux of nitrous oxide

On the basis of the methods described in paragraph 2, the nitrous oxide emission of the forest soil was measured in three series at the Nyírjes station in the Spring of 1997. In the majority of the installed cells emission can be detected with substantially high standard deviation (see *Fig. 2*). The average of the soil flux is  $2.0 \pm 0.8 \mu\text{gN m}^{-2} \text{ h}^{-1}$ . The atmospheric gradient of nitrous oxide at the last of the three sampling occasions was negative from the soil surface upwards prior to as well as following the gas sampling from the chambers. The values were  $-0.014 \text{ ppb N}_2\text{O m}^{-1}$  (n.s.) and  $-0.068 \text{ ppb N}_2\text{O m}^{-1}$  ( $p < 0.05$ ) when a linear model is used. This supports the conclusion that the soil emits nitrous oxide to the atmosphere as revealed by the chamber data.

Table 3. Summary of the dry deposition of nitrogen compounds at Nyírjes monitoring station

Period (quarter)	Dry deposition (emission) in mgN m <sup>-2</sup>							Total dry depos.
	Nitrous oxide	Nitric oxide	Nitrogen-dioxide	Ammonia	Nitric acid	Ammonium	Nitrate	
1st	(-19.3)	-14.7	-6.2	87.6	185.0	25.7	9.6	268
2nd	(-19.3)	-18.0	19.6	489.0	61.1	34.2	8.9	576
3rd	(-19.3)	-0.4	45.2	268.0	183.0	42.8	10.5	530
4th	(-19.3)	6.2	88.7	69.0	232.0	52.6	24.7	454
1996	-77.2 ?	-26.9	147.3	913.6	661.1	155.3	53.7	1 828

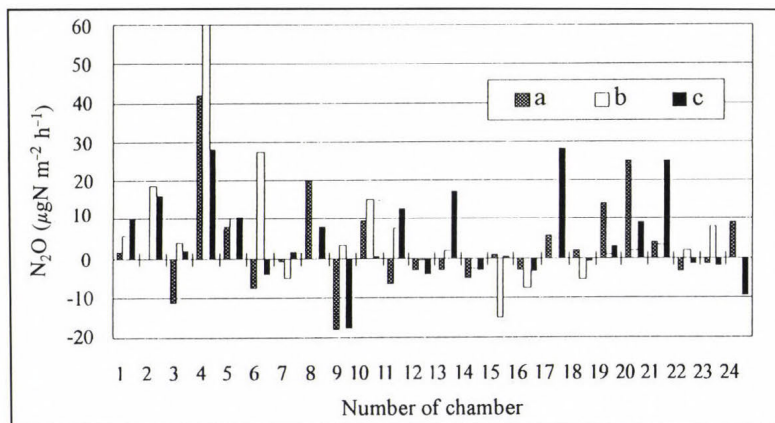


Fig. 2. Results of the three series (a, b, c) of measurements of dinitrogen oxide emission from the soil at Nyírjes station.

Because of the limited number of measurements the nitrous oxide emission for the whole year can not be estimated. The results concerning the dry spring period probably do not represent the yearly average. Therefore, the nitrous oxide emission has to be determined from figures from the literature. For spruce forest *Ambus* and *Christensen* (1995) found a 77 mgN m<sup>-2</sup> yr<sup>-1</sup> emission rate for nitrous oxide flux with a large spatial and temporal variation. This figure is 4.4 times higher (8.8 µgN m<sup>-2</sup> h<sup>-1</sup>) compared to the emission flux determined at Nyírjes station in Spring. In the calculation of the flux, as can be seen in Table 3, the data of the mentioned authors was used making the remark here that the estimation is somewhat uncertain because there could be differences between soil composition, humidity, etc. at the two places resulting in different emission rates.

### 3.4 Comparison of the determined dry deposition with the throughfall, stemflow deposition estimates

In dry periods the trace gases and particles are captured by the canopy partly on the surface of the leaves. The absorbed materials that are not taken up are leached by the following precipitation, which can be measured in the throughfall and in a small part in the stemflow samples. The nitrate and ammonium content generally increases, which can be measured in the wet only precipitation samples.

The dry deposition figures determined in paragraphs 3.1–3.3. are theoretically not comparable with the deposition estimated from the throughfall and stemflow measurements. One of the reasons is that the rate of net dry flux is the difference between deposition and emission terms. On the other hand dry deposition itself cannot be in relation with the deposition calculated by the throughfall estimation due to the following reason.

As mentioned, the flux of trace compounds can be characterised by the dry deposition velocity:

$$v_d(z) = \frac{-F_d}{C(z)}, \quad (2)$$

where  $v_d(z)$  and  $C(z)$  are the dry deposition velocity and concentration of the given compound at the height  $z$ , while  $F_d$  is the flux.

In the case of surfaces covered by vegetation, the dry deposition velocity can be described by the simple big leaf model as the result of the near surface resistances to the deposition. The effect of atmospheric turbulence can be characterised by the term  $r_a$ , as aerodynamic resistance. This term describes the rate of turbulent exchange processes i.e. the intensity of the turbulent transportation to the surfaces.

The turbulent processes are no longer effective from the displacement height. Below this in the so-called quasi-laminar layer, in the lack of regular turbulent motions, molecular diffusion and irregular motions transport the materials to the surface substantially slower than the turbulent motions. It is determined as  $r_b$  in the model.

The effect of the surface and the vegetation can be taken into account by the term  $r_c$ . This consists of different parts. When materials reach the level of leaves the uptake processes start immediately. Some of the materials like ozone, ammonia are mostly absorbed by stomata. This process can be described by  $r_{st}$  (stomatal) and  $r_{me}$  (mesophyll) resistances.

Other kinds of trace materials (like sulphur dioxide and nitric acid) are mostly adsorbed on the surface of the leaves (on the cuticle) remaining there until the following precipitation. This can be described as cuticular resistance,

$r_{cu}$ . Beside these processes the trace materials can be adsorbed on the trunk ( $r_{tr}$ ) and on the litter covered soil surface ( $r_{so}$ ). The rate of surface adsorption processes strongly depend on the wetness of the surface.

The stomatal uptake of materials depends on, beside the meteorological circumstances, the physiological condition of the plants i.e. the open or closed state of stomata. The rate reaches its maximum during the daytime hours of the vegetation period, but at higher temperatures and low relative humidity it decreases. There is a strong relation between the photosynthetic active radiation and the open state of stomata until a given value where the relation is inverse.

As a result of the different surface resistances the dry deposition velocity can be described by the following expression:

$$v_d(z) = \frac{1}{(r_a + r_b + r_c)}, \quad (3)$$

where

$$\frac{1}{r_c} = \frac{1}{(r_{st} + r_{me})} + \frac{1}{r_{cu}} + \frac{1}{r_{tr}} + \frac{1}{r_{so}}. \quad (4)$$

According to this theory, during the direct measurement of the dry deposition flux ( $F_d$ ) we have taken into consideration all of the terms of surface resistances  $r_a + r_b + r_c$ . However, in the dry deposition estimates calculated from the throughfall and stemflow figures the terms  $r_{st} + r_{me}$  and  $r_{so}$  are not represented.

The directly measured dry deposition rates and the dry deposition calculated from throughfall measurements are equal only in the case when these three latter terms are negligible. The deposition onto the soil is generally negligible, but stomatal uptake is dominant for a group of compounds, like carbon dioxide and ozone. As for nitrogen compounds the stomatal uptake of ammonia is dominant, but the effectiveness of this process can not be excluded for other kinds of nitrogen compounds, also.

There are some other reasons for the limited applicability of the estimation of dry deposition from throughfall results, namely the contamination of insects, according to a group of experts.

In spite of this we have attempted to compare the deposition figures calculated from the two methods, because from the difference of these figures we can estimate the magnitude of the stomatal uptake. On the other hand, because the dry deposition calculated directly ( $D_d$ ) is theoretically higher than the difference of throughfall and wet deposition (which is practically equal to  $D_d - D_{st}$ , where the latter term denotes the stomatal uptake) our theory is supported when the directly calculated deposition is higher.

The dry depositions calculated from throughfall and stemflow measurements are summarised in *Table 4*. The dry deposition without the stomatal uptake (and with the insect's contamination) can be seen in the last row. For the calculation of these figures the wet only deposition rate was subtracted from the sum of the throughfall and stemflow depositions. It seems from the data of the table that in the case of the nitrate ion, the substances leached from leaves cause only a 20 % increase compared to the wet deposition. For ammonium the increase is about 90 %, which is in agreement with the data in *Table 3*, where the deposition of ammonia and ammonium are much higher than that of nitric acid and nitrate. In the first quarter of the year the throughfall deposition gives a negative figure, which may be evidence for the limited applicability of throughfall estimations or indicates a possible nitrate and ammonium uptake from the precipitation by leaves through stomata.

*Table 4.* Wet deposition of nitrogen compounds and throughfall, stemflow data at Nyírjes monitoring station in the different quarters and during the whole year in mgN m<sup>-2</sup>

Kind of precipitation samples	Nitrate deposition					Ammonium deposition					Total N deposition
	1st	2nd	3rd	4th	1996	1st	2nd	3rd	4th	1996	1996
Wet only	96.9	123.0	92.4	79.4	391.7	93.3	216.0	144.0	69.5	522.8	915
Bulk throughfall	43.8	173.0	98.7	86.3	401.8	80.9	380.0	321.0	153.0	934.9	1 337
Stemflow	0	41.0	19.3	5.5	65.8	0	32.6	27.6	3.6	63.8	130
D <sub>d</sub> -D <sub>st</sub> -D <sub>w</sub>	-53.1	91.0	25.6	12.4	75.9	-12.4	196.6	204.6	87.1	475.9	552

(D<sub>d</sub>-D<sub>st</sub>-D<sub>w</sub>) is the dry deposition without the stomatal uptake (difference of the sum of throughfall + stemflow and wet only results).

The directly measured dry deposition figures (*Table 3*), in accordance with the minimum of the throughfall estimation, is at a minimum in the first quarter. In *Table 5* the directly measured dry deposition can be compared with the estimated deposition from the throughfall measurements. It follows from the table that on a yearly basis only a small part of the gross dry deposition (the emission part is not involved), i.e. 28.6 % can be detected in the throughfall samples. This suggests that the dry deposition of nitrogen compounds mostly takes place through the stomata. The cuticular and trunk uptake (adsorption) amounts to only a smaller part (29 %) of the total deposition. In the case when biological (insect) contamination is not negligible this rate is lower, i.e. stomatal uptake is more effective.

Table 5. The nitrogen balance between the atmosphere and a spruce forest in 1996 at Nyírjes monitoring station in  $\text{mgN m}^{-2}$

Period	Deposition		Total
	Dry	Wet	
1st quarter	268 (-66)	190	458
2nd quarter	576 (288)	339	915
3rd quarter	530 (231)	236	766
4th quarter	454 (100)	149	603
1996	1 828 (552)	914	2 742

In brackets are the dry deposition figures calculated from the difference of throughfall + stemflow and wet-only samples.

As a first approximation we may conclude that at least 71 % of the dry deposited nitrogen compounds,  $1\,380\text{ mgN m}^{-2}\text{ yr}^{-1}$  are taken up by stomata and can not be detected in throughfall samples.

In the case when nitrate and ammonium uptake from the precipitation by the stomata is also effective the stomatal uptake can be calculated from the difference of the sum of dry and wet deposition ( $2\,847\text{ mgN m}^{-2}\text{ yr}^{-1}$ ) and the throughfall + stemflow deposition ( $1\,467\text{ mgN m}^{-2}\text{ yr}^{-1}$ ) leading to the same figure,  $1\,380\text{ mgN m}^{-2}\text{ yr}^{-1}$ .

### 3.5. Estimation of the nitrogen balance

For the estimation of the nitrogen balance, the effects of the different deposition forms (dry + wet) need to be summarised. The dry + wet deposition for the year of 1996 and in the different quarters of the year together with the total deposition can be seen in Table 5. The dry deposition was calculated from the results of Table 3, while the wet deposition was estimated from the results of the wet only precipitation samples (Table 4). The table shows the net deposition figures also taking into account the emission.

From the results it can be concluded that in 1996, at the Nyírjes spruce forest, the nitrogen balance between the atmosphere and the forest ecosystem is  $2.7\text{ gN m}^{-2}\text{ yr}^{-1}$ , i.e. the different nitrogen compounds carry this amount of nitrogen to the forest from the atmosphere. Two thirds of the deposition is due to the dry deposition, which is higher than the previously estimated figure when lower deposition velocity figures (representative for other surfaces than forests) were applied (Horváth *et al.*, 1993a).

The reason for the difference can be explained by the fact that surface roughness for the forest is higher compared with other surfaces resulting in

higher turbulent exchange as a consequence of the lower aerodynamical resistance ( $r_a$ ). The canopy resistance  $r_c$  is also lower for forests because of the relatively dense surface characterised by the leaf area index.

Furthermore, it can be concluded that the emission processes from the soil are not in competition with the deposition, the nitrogen oxides emitted from the forest soil transport only a few percent of the deposited nitrogen into the atmosphere.

The nitrogen balance determined here has some uncertainty. The calculation of nitrous oxide emission was estimated by literature figures, which will probably will be modified with the results of measurement being carried out at present. The direct measurements of the dry deposition velocities of ammonia, nitric acid and ammonium, nitrate particles will also be required at Nyírjes station to avoid the error in estimation caused using literature data concerning forests with different climatic circumstances.

Hopefully, these investigations will be fulfilled in the future, but it seems from these results that the nitrogen balance will not differ substantially from the figures determined in this work.

The  $2.7 \text{ gN m}^{-2} \text{ yr}^{-1}$  deposition figure refers to a  $27 \text{ kgN ha}^{-1} \text{ yr}^{-1}$  rate, which is not negligible from the point of view of the forest ecosystem. It is probable that one half of the deposited nitrogen compounds are taken up by stomata partly as nutrients and partly causing damaging effects. The other half of the nitrogen compounds leach into the forest soil where they partly accumulate and partly run off with the soil water. To determine this division complex air chemistry, forestry and hydrological investigations are needed.

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## The Penman-Monteith concept based land-surface model PMSURF

Ferenc Ács<sup>1</sup> and Michael Hantel<sup>2</sup>

<sup>1</sup>*Department of Meteorology, Eötvös Loránd University,  
H-1117 Budapest, Pázmány Péter sétány 1/a, Hungary,  
e-mail: acs@nimbus.elte.hu*

<sup>2</sup>*University of Vienna, Hohe Warte 38, A-1190 Vienna, Austria*

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**Abstract**—The land-surface model PMSURF, designed jointly at the Universities of Vienna and Budapest, is reviewed. PMSURF consists of one canopy layer and three soil layers. Prognosed state quantities are: stored water in the vegetation layer and soil moisture content in the soil layers. The forecast equations for these variables rest upon the budget laws for water substance. There is no temperature prediction, therefore net radiation or surface temperature is needed as input. This fact implies that in winter application snow and soil freezing/melting processes cannot be represented. Fluxes in the water equations comprise rain and rain interception, canopy drainage and infiltration, surface runoff, soil water diffusion, root water uptake, evapotranspiration, conductance of water through roots and stems and subsurface runoff. Energy and water fluxes are parameterized with the resistance concept.

PMSURF is tested on off-line mode for the Cabauw data set. The observed annual mean values and the seasonal changes of turbulent and water fluxes are satisfactorily reproduced. For example, the model yields latent and sensible heat fluxes of  $-38.0$  and  $-4.1$   $\text{W/m}^2$ , evapotranspiration and runoff of  $-482$  and  $291$   $\text{mm/year}$ .

The sensitivity of model performance to the choice of turbulent heat flux parameterization concept is also studied by comparing PMSURF to the land-surface model PROGSURF (Ács and Hantel, 1998b). Sensitivity tests enable us to establish and to quantify the link between the model performance and the responsible mechanisms of turbulent heat flux parameterization concepts. The main results are as follows: PMSURF-calculated turbulent heat and water fluxes are somewhat closer to the observations than those calculated by PROGSURF. Further the course of evapotranspiration/soil moisture curve does not depend upon the turbulent heat flux parameterization concept.

The results are useful in optimizing evapotranspiration parameterization in land surface models. PMSURF presently serves to simulate observed surface fluxes for an atmospheric diagnostic model.

**Key-words:** parameterization of land-surface processes, turbulent heat parameterization concept, intercomparison of models.

## 1. Introduction

The *Project for Intercomparison of Land-surface Parameterization Schemes* (PILPS), which is part of the *World Climate Research Programme* (WCRP) under the auspices of the *Global Energy and Water Cycle Experiment* (GEWEX) and the *Working Group on Numerical Experimentation* (WGNE) tends to improve the understanding of land-surface models (LSM) with respect to their differences. The differences appear on three levels; according to this they can be classified into three categories:

- (1) Differences in the parameterization of the parameters. The relevant parameters are radiation properties (reflexion, transmission and absorption) and all resistances and/or conductances in the soil-plant-atmosphere system. Among them the albedo and the surface resistances seem to be more important.
- (2) Differences in the parameterization of the fluxes. The turbulent heat fluxes can be parameterized either by the Penman-Monteith's concept or by gradient laws. There are also many conceptual differences in the parameterization of water balance components. For instance, the transpiration can be parameterized by using three different concepts (Mahfouf *et al.*, 1996): the supply-demand approach, the canopy surface resistance concept and the moisture availability concept (ratio of actual and potential evapotranspiration). There are also basic differences in runoff parameterizations (Wetzel *et al.*, 1996).
- (3) Differences in the application of prognostic equations. The type and the number of prognostic equations determine the model structure. The equations and their numerical implementation into the model can largely differ from model to model. These differences are often called as structural differences.

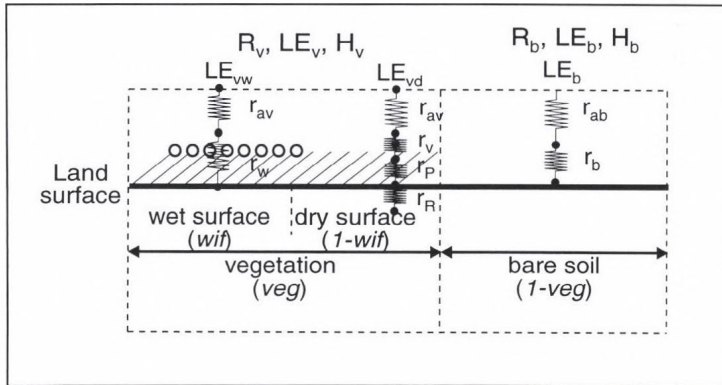
LSM with different structure, flux and/or parameter parameterizations were compared under diverse climatic conditions, e.g., at Caumont in Southern France (Henderson-Sellers, 1996), at Cabauw in the Netherlands (Chen *et al.*, 1997), or at Valdai in Russia (Schlosser *et al.*, 1997). These studies showed that there was considerable scatter between the PILPS models; further it was impossible to establish a link between the model's performance and the responsible mechanisms because most LSM differed in various aspects simultaneously (Shao and Henderson-Sellers, 1996).

The objective of this study is therefore twofold. First, we describe and validate the land-surface model PMSURF. The validation is performed using Cabauw data set. Second, we shall address the sensitivity issue studying and comparing model runs obtained by PMSURF and PROGSURF. PMSURF and PROGSURF models differ only in the parameterization of turbulent heat fluxes; so the sensitivity test reveals and quantifies the impact of turbulent heat flux parameterization concepts upon model performance.

## 2. Model

PMSURF (**P**enman-**M**onteith's **S**urface **F**luxes) is based on previous work of *Ács* (1995, 1996) and *Ács* and *Hantel* (1998a, 1998b). It comprises one vegetation layer and three soil layers. The core of the scheme is a 3-layer diffusion type soil moisture prediction (*Sellers et al.*, 1986) in combination with *Penman-Monteith* (PM) concept for turbulent heat flux calculation. This means that there is no temperature prediction module, therefore net radiation or surface temperature is needed as input for its application. PMSURF is similar to *Dolman's* (1993) model; its structure differs from SSiB-type models (e.g., *Ács*, 1994; *Xue et al.*, 1996) in the sense that PMSURF is designed to require only a minimum of soil vegetation parameters as input. Sub-gridscale variations of surface characteristics are not considered here.

The transport processes are quantified through the resistance formalism. *Fig. 1* shows the resistance system as implemented in PMSURF. The land is subdivided into a vegetated (*veg*) and a non-vegetated (bare soil) part ( $1 - veg$ ). The vegetated land (terms *vegetation* and *canopy* will be used synonymously) is again subdivided into a wet vegetation (*wif*) and a dry vegetation part ( $1 - wif$ ). Each of these subtypes is individually homogeneous. The specific surface characteristics are expressed via aerodynamic and surface resistances. In PMSURF we follow the convention to count all vertical fluxes positive if directed downwards. The consequence is that, e.g., evaporation and transpiration are practically always negative.



*Fig. 1.* The resistance system as implemented in PMSURF. Symbols:  $R_v$ ,  $H_v$ ,  $LE_v$  = net radiation, latent and sensible heat fluxes above vegetation;  $LE_{vw}$  and  $LE_{vd}$  = latent heat flux above wet and dry vegetation;  $R_b$ ,  $H_b$ ,  $LE_b$ : as before but for bare soil;  $r_{av}$  and  $r_{ab}$  = aerodynamic resistance above vegetation and bare soil;  $r_w$ ,  $r_v$ ,  $r_b$  = surface resistance of wet vegetation, dry vegetation, bare soil;  $r_p$  = plant resistance,  $r_R$  = soil resistance in root zone, *wif* = wet part of canopy surface; *veg* = vegetated part of land-surface.

In the following those parts of PMSURF are presented which deviate from PROGSURF. These are: the prognostic equations, the heat flux parameterization and the leaf water potential representation.

## 2.1 Prognostic equations

The model has four prognostic variables: intercepted water stored in vegetation layer  $M_v$  and soil moisture content in the 1st, 2nd and 3rd soil layer  $\theta_i$ , for  $i = 1, 2, 3$ .

Water storage in the vegetation layer is predicted by:

$$\frac{\partial M_v}{\partial t} = P_v - D_v + E_{vw}, \quad (2.1)$$

where  $P_v$  is the interception of water by vegetation,  $D_v$  is the drainage of water from vegetation and  $E_{vw}$  is evaporation from wet parts of the vegetation ( $E_{vw} < 0$ ) or dew formation ( $E_{vw} > 0$ ).

Diffusion-type moisture prediction is applied in the soil layers. The prognostic equations for the three layers are:

$$\rho_w \cdot D_1 \cdot \frac{\partial \theta_1}{\partial t} = P_{inf} - Q_1 + Q_{r0} - Q_{r1} + E_b - Q_{run1}, \quad (2.2)$$

$$\rho_w \cdot D_2 \cdot \frac{\partial \theta_2}{\partial t} = Q_1 - Q_2 + Q_{r1} - Q_{run2}, \quad (2.3)$$

$$\rho_w \cdot D_3 \cdot \frac{\partial \theta_3}{\partial t} = Q_2 - Q_3 - Q_{run3}, \quad (2.4)$$

where  $\rho_w$  is water density,  $D_i$  is the depth of the  $i$ th soil layer,  $\theta_i$  is the soil moisture content in the  $i$ th soil layer,  $P_{inf}$  is the water infiltrated into the soil,  $E_b$  is the evaporation from bare soil,  $Q_{r0}$  and  $Q_{r1}$  is the root water flux across surface and across bottom of the 1st soil layer,  $Q_1$  and  $Q_2$  is the water diffusion between adjacent layers,  $Q_3$  is the gravitational drainage and  $Q_{runi}$  represents the lateral drainage from the  $i$ th layer. The gravitational and lateral drainage terms are positive representing outflow from the system.

## 2.2 Heat fluxes

The latent heat flux is parameterized using Penman-Monteith concept (Monteith, 1965):

$$L \cdot E_j = \frac{\Delta \cdot A_j + \rho c_p \delta e / r_{aj}}{\Delta + \gamma (1 + r_j / r_{aj})}, \quad (2.5)$$

where  $L$  is the latent heat of vaporization,  $\Delta$  is the slope of saturated vapor pressure curve at reference temperature  $T_r$ ,  $A_j$  is the available energy of surface,  $\rho$  is the air density,  $c_p$  is the specific heat of air at constant pressure.  $\delta e = e_s(T_r) - e_r$  is the vapor pressure deficit,  $e_s(T_r)$  is the saturation vapor pressure at  $T_r$  and  $e_r$  is the vapor pressure at reference level.  $\gamma$  is the psychrometric constant,  $r_j$  and  $r_{aj}$  are the surface and aerodynamic resistance, respectively. The additional index  $j$  refers to the domains of vegetation ( $j = v$ ) with relative coverage  $veg$ , and of bare soil with coverage  $1 - veg$  ( $j = b$ , see Fig. 1). For bare soil  $A_j = R_j - G_j$  but for vegetation  $A_j = R_j$ .  $R_j$  is the net radiation flux while  $G_j$  is the soil surface heat flux. The horizontal mean latent heat flux is:

$$L \cdot E \quad \text{with} \quad E = veg \cdot E_v + (1 - veg) \cdot E_b. \quad (2.6)$$

For vegetation we additionally distinguish between wet ( $j = vw$ ) and dry ( $j = vd$ ). The wet/dry distinction applies only to the surface resistance  $r_j$ . Thus  $E_v = wif \cdot E_{vw} + (1 - wif) \cdot E_{vd}$ . For simplicity, the vegetation resistances  $r_{vw}$ ,  $r_{vd}$  will be abbreviated as  $r_w$ ,  $r_v$ , respectively.  $wif$  is parameterized after *Sellers et al.* (1986).

The sensible heat flux is estimated as residual,

$$H_j = A_j - L \cdot E_j. \quad (2.7)$$

The weighted mean value is:

$$H = veg \cdot H_v + (1 - veg) \cdot H_b. \quad (2.8)$$

The soil surface heat flux under canopy  $G_v$  is assumed to be zero,

$$G_v = 0. \quad (2.9)$$

The bare soil heat flux is empirically estimated after *Nickerson and Smiley* (1975):

$$G_b = 0.15 \cdot R_b. \quad (2.10)$$

The horizontal mean soil heat flux is:

$$G = veg \cdot G_v + (1 - veg) \cdot G_b. \quad (2.11)$$

### 2.3 Calculation of leaf water potential

The leaf water potential  $\Psi_v$  is calculated using water flow continuity assumption in the soil-plant-atmosphere system; that is we suppose that the transpiration is equal to the root water uptake. Combining formulae for transpiration, root water uptake, canopy resistance and moisture availability, it is possible to get a quadratic equation for  $\Psi_v$  (see the Appendix):

$$a \cdot \Psi_v^2 + b \cdot \Psi_v + c = 0. \quad (2.12)$$

The coefficients  $a$ ,  $b$  and  $c$  are given in the Appendix. Analyzing the order of magnitude of the quantities, it is possible to show that only one solution of the quadratic equation is physically based. The physically correct solution of  $\Psi_v$  is obtained by:

$$\Psi_v = \frac{-b + [b^2 - 4 \cdot a \cdot c]^{1/2}}{2a}. \quad (2.13)$$

### 2.4 Numerical implementation of the model

The sequence of calculations in a given time step is as follows: First, the radiation module calculates the net radiation of land-surface estimating separately the albedo for bare soil and vegetation. For the Cabauw data set according to PILPS 2a specifications, the vegetation and bare soil albedo is constant and therefore the corresponding albedo subroutines are not used.

Heat and water flux calculations follow the radiation module. The *vegetation module* calculates turbulent heat fluxes above vegetation, root water fluxes and water fluxes in vegetation layer. The *bare soil module* contains turbulent and soil heat flux parameterization above bare soil. In case of unstable stratification the flux/aerodynamic resistance relationship is iteratively calculated for both modules. The *ground module* calculates the soil heat flux of vegetation-ground system as weighted mean of its vegetated and non-vegetated components. Infiltration, surface runoff, soil water diffusion, lateral runoff and gravitational drainage are calculated without making such distinction.

At the end the prognostic equations (water and soil moisture storages in the vegetation and soil layers) are applied in a separate module. They are solved using an explicit time scheme. The time step used was 900 s.

### 3. Model validation

PMSURF has been extensively tested in off-line mode using the 1987 data from Cabauw, Netherlands. The well known dataset has been described and analyzed in detail by *Beljaars and Bosveld* (1997) and it is also used in PILPS project, phase 2a. In the numerical experiments PMSURF was always initialized as all PILPS participating models by saturating all liquid water stores. The variable and constant land-surface parameters are specified according to the specifications used in PILPS, 2a experiment (see Table A2, A3 and A4 in *Chen et al.*, 1997).

The model validation is performed by comparing simulated and observed surface fluxes. Among the results, the annual mean characteristics and seasonal changes of some heat and water balance components are presented and analyzed.

#### 3.1 Annual mean characteristics and seasonal variations

The spinup time of the PMSURF model is 2 years, that is the equilibrium year is achieved in the 2nd year. The annual mean characteristics refer to the equilibrium year.

The annual mean sensible and latent heat fluxes obtained by PMSURF1 (standard PMSURF run) are presented on *Fig. 2* together with the other PILPS results (for details see *Fig. 5* in *Chen et al.*, 1997). The results of PMSURF2 and PROGSURF will be discussed in Section 4. The sensible heat flux of PMSURF1 is  $-4.1 \text{ W/m}^2$ , the latent heat flux is  $-38 \text{ W/m}^2$ . The corresponding point on *Fig. 2* is exactly located on the net radiation line; this is clear because the measured net radiation is used as input.

The annual runoff versus evapotranspiration is given on *Fig. 3*. (The figure is worth to compare to *Fig. 10* in *Chen et al.*, 1997.) PMSURF1 yields  $-482 \text{ mm/year}$  evapotranspiration and  $291 \text{ mm/year}$  runoff, while the corresponding observed fluxes were  $-525$  and  $250 \text{ mm/year}$ . The annual mean soil water in the root zone obtained by PMSURF1 is  $340 \text{ mm}$ . Its estimated value (indirectly observed) is about  $350 \text{ mm}$  (*Chen et al.*, 1997).

The seasonal change of latent and sensible heat fluxes is presented on *Fig. 4*. The largest deviation between simulated and observed  $L \cdot E$  values is about  $15 \text{ W/m}^2$  and it appears in March and May. The largest deviation for  $H$  is about  $10 \text{ W/m}^2$  and it appears in April and May.

PMSURF's performance can be understood in great extent analysing the evapotranspiration/soil moisture relationship  $E(\theta)$  (for details see Section 4.3). Qualitatively, the two governing parameters of the  $E(\theta)$ -curve are the slope  $S = \partial E(\theta)/\partial \theta$  in the transition region (i.e., region between water stressed and well watered conditions) and the saturation value  $E(\theta_S)$ ; at Cabauw  $\theta_S = 0.468 \text{ m}^3/\text{m}^3$ . *Ács and Hantel* (1998a) showed that  $S$  is controlled by the

parameterization of  $F_{ma}$ , while  $E(\theta_S)$  is controlled by the parameterization of  $F_{ad}$  (see Eq. (A.4)). They also showed that the evapotranspiration for the Cabauw data set is predominantly controlled by the parameter  $E(\theta_S)$ ; the parameter  $S$  is of minor influence at the Cabauw site.

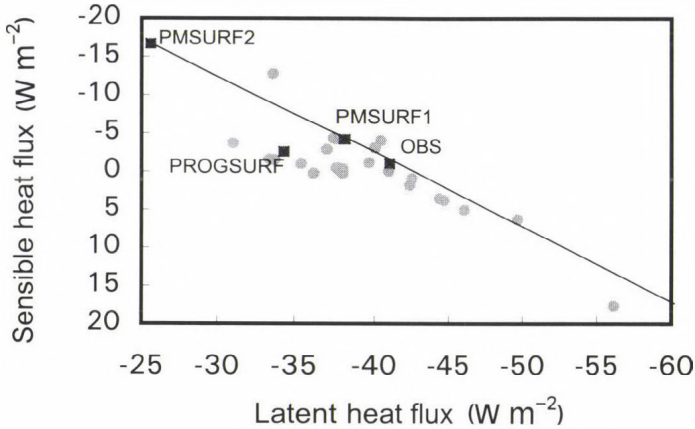


Fig. 2. Annually averaged sensible versus latent heat fluxes estimated by the different versions of PMSURF and PROGSURF (thick symbols) along with the equivalent PILPS phase 2a results (thin dots). “PMSURF1” = standard PMSURF run; “PMSURF2” = PMSURF run with latent heat flux parameterized by gradient formula “PROGSURF” = standard PROGSURF run; and “OBS” = observed value.

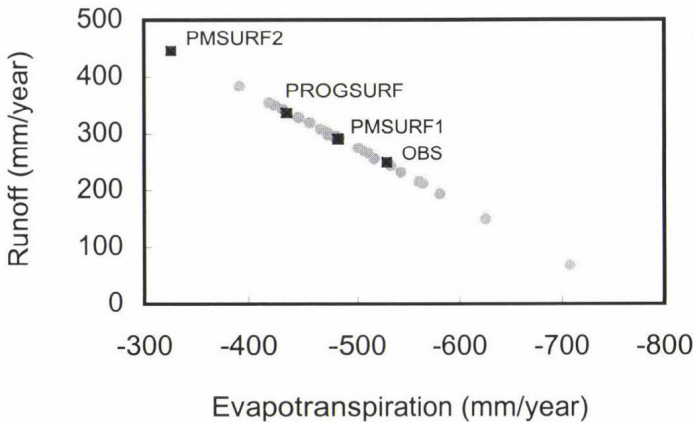


Fig. 3. Annual runoff versus evapotranspiration.

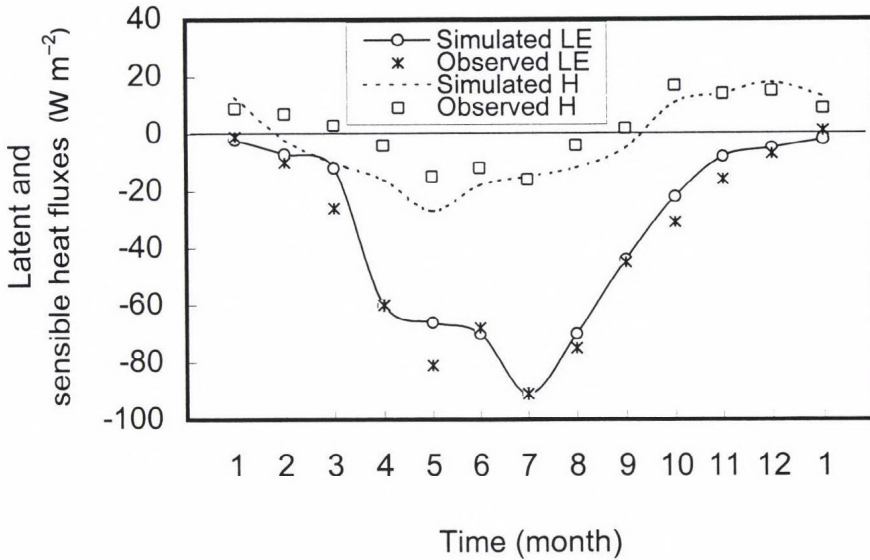


Fig. 4. Annual course of latent and sensible heat fluxes simulated by standard PMSURF.

#### 4. Sensitivity tests

The sensitivity of model performance to the choice of turbulent heat flux parameterization concept is analysed. The sensitivity tests are carried out comparing two different modes of the PMSURF and the PROGSURF model.

In PMSURF the turbulent heat fluxes are parameterized using Penman-Monteith's concept. This implies the parameterization of soil and latent heat fluxes and the determination of sensible heat flux as the residual term from the energy balance equation. Therefore there is no surface temperature prediction, so net radiation and/or surface temperature are required as input. Soil heat flux is usually parameterized as the percentage of net radiation. The latent heat flux can be parameterized either by Penman-Monteith's combination equation (this is the common case) or by gradient formula. The former calculation mode for  $L \cdot E$  represents the standard PMSURF referred to as PMSURF1. The latter calculation mode of  $L \cdot E$  is not commonly used and it is referred to as PMSURF2.

In PROGSURF the turbulent heat flux parameterization is based on gradient law, that is latent and sensible heat fluxes are parameterized via gradient formulae independently from each other. This implies the soil surface temperature prediction, the parameterization of net radiation and the calculation of soil heat flux via heat conduction equation.

It is obvious that the two turbulent heat flux parameterization concepts are completely different. In PMSURF-approach the turbulent heat fluxes are diagnostically determined; whereas in PROGSURF-approach they are determined via gradient formulae which implies soil surface temperature prediction. But since there is no any other difference between the models, they can also be treated as two different modes of one model. Using this model architecture it is possible to study the link between the model's performance and the responsible mechanisms. So the PMSURF1/PMSURF2 comparison shows the deviations produced by the differences in the parameterization of latent heat flux (Penman-Monteith equation versus aerodynamic formula). The PMSURF2/PROGSURF comparison reveals the deviations in model performance produced by the application of force-restore method, while the PMSURF1/PROGSURF comparison quantifies the deviations produced by the differences in the turbulent heat flux parameterization concepts.

#### *4.1 Annual mean characteristics*

The annual mean sensible and latent heat fluxes obtained by PMSURF1, PMSURF2 and PROGSURF just introduced and the land-surface models participating in PILPS phase 2a have been presented on Fig. 2. The points of PMSURF1 and PMSURF2 lay on the observed net radiation line. This is clear since PMSURF uses observed radiation surface temperature as input. PMSURF1 is much more closer to the observation than PMSURF2. The PMSURF1/PMSURF2 deviation is very large. This deviation is caused by the differences in the parameterization of the latent heat flux. The Penman-Monteith-based latent heat flux parameterization (PMSURF1) uses indirectly the radiation surface temperature via net radiation. In spite of this, gradient formula based latent heat flux parameterization (PMSURF2) depends directly upon the radiation surface temperature. Obviously, the observed radiation surface temperatures seem to be too small, in the sense that the latent heat flux calculated by gradient formula ( $25.6 \text{ W/m}^2$ ) is far underestimated with respect to its observed value (about  $41 \text{ W/m}^2$ ).

The PROGSURF-point deviates from the observed net radiation line. This indicates an error in the prediction of vegetation-ground temperature  $T_{vg}$ . The yearly mean of  $T_{vg}$  temperature ( $281.6 \text{ K}$ ) is somewhat overestimated with respect to the radiation surface temperature ( $280.7 \text{ K}$ ). This considerably improves the sensible and latent heat flux calculation but slightly underestimates the net radiation (the yearly mean of  $R$  is  $37.9 \text{ W/m}^2$  instead of  $42.3 \text{ W/m}^2$ ) as compared to PMSURF2. The PMSURF2/PROGSURF comparison shows the effect of force-restore method upon model performance. In spite of this, the PMSURF1/PROGSURF comparison indicates the effect of turbulent heat flux parameterization concept upon model performance. This effect seems to be not too significant though the results of PMSURF1 are in better agreement with observations than those of PROGSURF.

The annual runoff versus evapotranspiration has been presented on Fig. 3. The PMSURF1/PMSURF2 deviation caused by latent heat flux parameterization differences is the greatest. The difference in the evapotranspiration is about 160 mm. According to PMSURF2-results the observed radiation surface temperatures seem to be too small. The PMSURF2/PROGSURF deviation is great, but somewhat smaller than the PMSURF1/PMSURF2 deviation. The PROGSURF-results are much closer to the observation due to application of the force-restore method. The PMSURF1/PROGSURF deviation is the smallest. The difference in the evapotranspiration is about 50 mm. The PMSURF1-results are closer to the observation than the PROGSURF-results; in this sense the approach based on Penman-Monteith's concept seems to be more advantageous than the approach based on the gradient law.

#### 4.2 Seasonal changes of energy and water balance components

Seasonal changes of soil heat flux obtained by PMSURF1, PMSURF2 and PROGSURF are presented on Fig. 5. The parameterization of soil surface heat flux in PMSURF1 and PMSURF2 is identical (see Eqs. (2.9) to (2.11)), so there is no difference between their courses. The course is unreal, it is represented by a constant close to zero. This fact is of no importance because

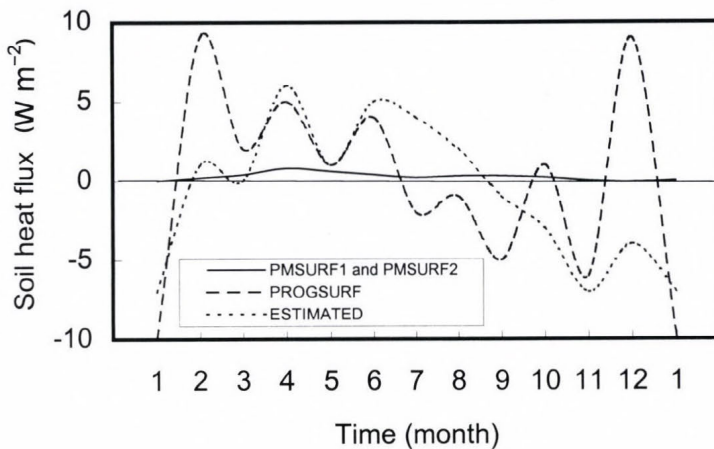


Fig. 5. Annual course of soil heat flux simulated by different modes of PMSURF and PROGSURF.

$G$  is the least surface energy balance term. The estimated  $G$  on the basis of measured soil heat flux at 5 and 10 cm depth (Beljaars and Bosveld, 1997) is

also small (not greater than  $8 \text{ W/m}^2$ ) but it shows a yearly course. In PROGSURF the soil heat flux at 10 cm depth is parameterized by the heat conduction equation in conjunction with the force-restore method. The course obtained is much more real than PMSURF's course.

The annual course of evapotranspiration  $E$  obtained by the PMSURF modes and PROGSURF is presented on Fig. 6. The figure pictures strong seasonal changes of evapotranspiration. Note that at the specific site of Cabauw,  $veg$  is quite close to unity, i.e., evapotranspiration is practically equal to transpiration. PMSURF1, PROGSURF and PMSURF2 shows the greatest, the medium and the smallest annual amplitude, respectively. Both the PMSURF and the PROGSURF underestimate the absolute value of evapotranspiration with respect to observation. The greatest deviations of evapotranspiration are between PMSURF1 and PMSURF2. They are most pronounced in summer with maximum of almost 20 mm in July. The performance of PROGSURF is between PMSURF1 and PMSURF2, but much closer to PMSURF1.

The annual course of modeled total runoff for the PMSURF and PROGSURF runs (observations do not exist) is reproduced on Fig. 7. The greatest runoff is produced by PMSURF2. It changes between 40 and 50 mm/month in winter; in summer it is much smaller, but still greater than 10 mm/month.  $Q_{run}$  courses obtained by PMSURF1 and PROGSURF are very similar and close to each other. The greatest deviation between them appears in June and it amounts to about 10 mm/month. According to these runs there is no runoff in July and August, which is in accordance with the results of *Chen et al.* (1997).

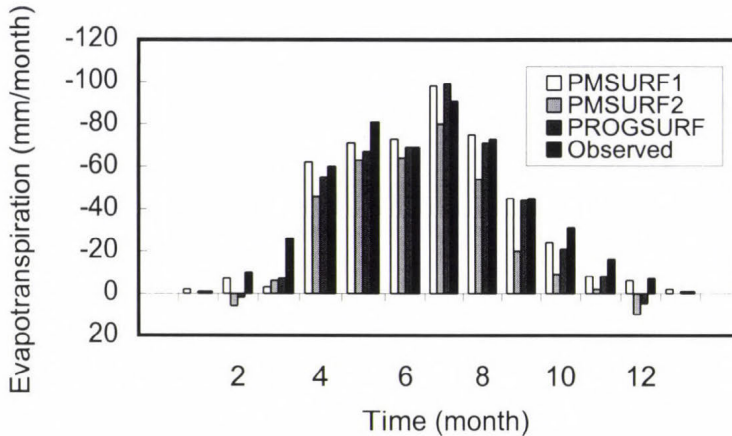


Fig. 6. Annual course of evapotranspiration simulated by different modes of PMSURF and PROGSURF.

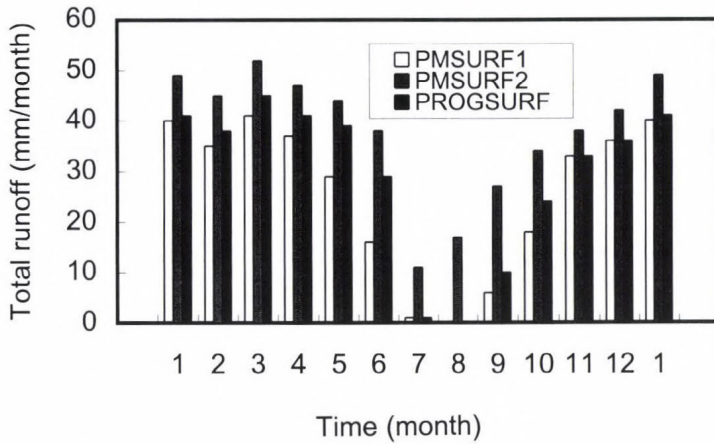


Fig. 7. Annual course of total runoff simulated by the different modes of PMSURF and PROGSURF.

#### 4.3 Evapotranspiration/soil moisture relationship

The evapotranspiration/soil moisture relationship is a fundamental relationship in each model. It is determined by many factors; in this sense it can also be determined by the turbulent heat flux parameterization concept. Here this fact will be briefly controlled and discussed.

The  $E(\theta)$  function (including the factor  $L$ ) for standard PMSURF and PROGSURF is drawn on Fig. 8. The curves are obtained using the following data. Soil vegetation parameters are from the Cabauw data set. The vegetation-ground temperature used in PMSURF as input is the equilibrium temperature estimated by PROGSURF solving the energy balance equation of vegetation-ground system. In both runs the following atmospheric conditions were kept fixed: global radiation =  $800 \text{ W/m}^2$ , air temperature =  $25.8^\circ\text{C}$ , vapor pressure =  $18 \text{ hPa}$ , wind velocity =  $6.0 \text{ m/s}$  and precipitation =  $0 \text{ mm}$ . The height of reference level is  $20 \text{ m}$ .

There is practically no difference between the  $E(\theta)$  curves obtained by the PMSURF1 and PROGSURF. This means that the turbulent heat flux parameterization concept does not affect the course of  $E(\theta)$  curve when all other parameterizations and conditions are identical in PMSURF1 and PROGSURF. All these results and analyses suggest that at the Cabauw site the surface radiation temperatures seem to be too small, in the sense that the latent heat flux calculated by gradient formula using surface radiation temperature is far underestimated with respect to its observed values.

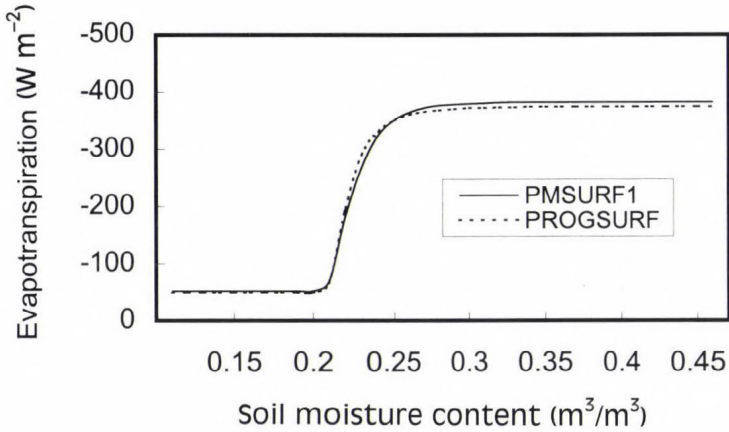


Fig. 8. Evapotranspiration versus root zone soil moisture simulated by the standard PMSURF and PROGSURF.

### 5. Conclusion

The land-surface model PMSURF has been briefly documented with respect to the land-surface model PROGSURF (Ács and Hantel, 1998a). PMSURF deviates from the PROGSURF as follows:

- (1) net radiation or surface temperature is used as input,
- (2) soil surface heat flux is calculated via net radiation,
- (3) latent heat flux is parameterized via Penman-Monteith's combination equation (Monteith, 1965). This representation is implemented in both the aerodynamic resistance and leaf water potential calculations,
- (4) sensible heat flux is estimated as the residual from the energy balance equation. These changes imply that in winter application snow and soil freezing/melting processes cannot be represented.

PMSURF has been tested in off-line mode for the Cabauw data set, using the same specifications that have been applied in the PILPS campaign (Chen *et al.*, 1997). The model reproduces satisfactorily both the observed annual mean values and the seasonal changes of turbulent and water fluxes. For example, the annual mean values of evapotranspiration and runoff are -482 and 291 mm, respectively. Nevertheless, soil surface heat flux is not satisfactorily reproduced.

The model sensitivity to the choice of turbulent heat flux parameterization concept has also been studied by comparing the standard PMSURF (referred to as PMSURF1), its modified version (referred to as PMSURF2) and the PROGSURF. The comparative analyses enable us to establish and to quantify the link between the model performance and the responsible mechanisms of turbulent heat flux parameterization concepts. The results can be summarized as follows:

- (1) The PMSURF1/PMSURF2 comparison shows the model performance deviations produced by the differences in the parameterization of latent heat flux (Penman-Monteith equation versus aerodynamic formula). PMSURF2 underestimates significantly the evapotranspiration with respect to the observations and the simulation results of PMSURF1. This result suggests that the observed surface radiation temperatures seem to be too small to be successfully used in gradient formula.
- (2) The PMSURF2/PROGSURF comparison reveals the deviations produced by the application of force-restore method. PMSURF2/PROGSURF comparison shows that the application of force-restore method improves considerably the turbulent heat flux calculation overestimating  $T_{vg}$  temperatures with respect to the observed temperatures. Note that the observed temperature is surface radiation temperature and the  $T_{vg}$  temperature is the equilibrium temperature of the vegetation-ground system.
- (3) PMSURF1/PROGSURF comparison quantifies the deviations produced by the differences in the turbulent heat flux parameterization concepts. PMSURF1-calculated turbulent heat and water fluxes are somewhat closer to the observations than those calculated by PROGSURF. Further the course of  $L \cdot E(\theta)$  curve does not depend upon turbulent heat flux parameterization concept.

This series of experiments allow us to qualify PMSURF as a PILPS-tested model (*Henderson-Sellers et al.*, 1993). It is hoped that the further inter-comparison campaigns presently in preparation may be useful in optimizing evapotranspiration parameterization in land surface schemes. PMSURF is presently used to specify the boundary conditions (as substitute for observed fluxes of latent and sensible heat) for the software DIAMOD (*Hantel et al.*, 1993; *Haimberger et al.*, 1995) which is routinely used at the University of Vienna to diagnose the convective fluxes in the free atmosphere.

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## APPENDIX

### Parameterization of $\Psi_v$ in PMSURF

Water transfer through plants is characterised by the transpiration and root water uptake. Both fluxes are parameterized using a resistance formalism in the soil-plant-atmosphere system:

$$E_{vd} = -\frac{1}{L} \cdot \frac{\Delta \cdot R_v + \rho c_p [e_s(T_r) - e_r]/r_{av}}{\Delta + \gamma(1 + r_v/r_{av})} \quad (\text{A.1})$$

and

$$Q_{r0} = -\rho_w \frac{(\Psi_R - z_T) - \Psi_v}{r_R + r_P}, \quad (\text{A.2})$$

where  $\rho_w$  is the water density,  $\Psi_R$  is the soil moisture potential in the root zone and  $z_T$  is the vegetation source/sink height. It is well known, that the water storage in the plants is negligible with respect to water in- and outfluxes, so that it is reasonable to use a water flow continuity assumption (Rutter, 1975):

$$Q_{r0} = veg \cdot (1 - wif) \cdot E_{vd}. \quad (\text{A.3})$$

At the same time both fluxes depend on leaf water potential  $\Psi_v$ :  $Q_{r0}$  explicitly, while  $E_{vd}$  implicitly via canopy resistance  $r_v$ .  $r_v$  is expressed through a multiplicative formula as follows:

$$r_v = \frac{r_{stmin} \cdot F_{ad}}{LAI \cdot GLF \cdot F_{ma}} \quad (\text{A.4})$$

with

$$F_{ma} = \frac{\Psi_v - \Psi_{cr}}{\Psi_{SR} - \Psi_{cr}}. \quad (\text{A.5})$$

Note that the notation used is as in *Ács* and *Hantel* (1998b). We see, that there are five equation with five unknowns:  $E_{vd}$ ,  $Q_{r0}$ ,  $r_v$ ,  $F_{ma}$  and  $\Psi_v$ . All other terms either are constants or can be expressed independently from  $\Psi_v$ .  $\Psi_v$  is possible to get starting from Eq. (A.3). After some rearranging,

$$[(\Psi_R - z_T) - \Psi_v] \cdot [r_{av} \cdot (\Delta + \gamma) + \gamma \cdot r_v] \cdot L \cdot \rho_w - RSP = 0, \quad (\text{A.6})$$

where  $RSP$  is independent from  $\Psi_v$ , defined as

$$RSP = veg \cdot (1 - wif) \cdot \Delta \cdot R_v \cdot r_{av} \cdot (r_R + r_P) + \rho c_p [e_s(T_r) - e_r] \cdot (r_R + r_P). \quad (\text{A.7})$$

Replacing  $r_v$ ,  $F_{ma}$  and introducing two new variables independent from  $\Psi_v$ , the Eq. (A.6) turns into the following equation:

$$[(\Psi_R - z_T) - \Psi_v] \cdot \left[ AC + MF \cdot \frac{\Psi_{SR} - \Psi_{cr}}{\Psi_v - \Psi_{cr}} \right] - RSP = 0, \quad (\text{A.8})$$

with

$$AC = r_{av} \cdot (\Delta + \gamma) \cdot L \cdot \rho_w \quad (\text{A.9})$$

and

$$MF = \gamma \cdot \frac{r_{stmin} \cdot F_{ad}}{LAI \cdot GLF} \cdot L \cdot \rho_w. \quad (\text{A.10})$$

Finally, multiplying Eq. (A.8) by  $\Psi_v - \Psi_{cr}$  and rearranging it, we can get a quadratic equation for  $\Psi_v$ :

$$a \cdot \Psi_v^2 + b \cdot \Psi_v + c = 0, \quad (\text{A.11})$$

where

$$a = 1, \quad (\text{A.12})$$

$$b = -(\Psi_R - z_T) - \Psi_{cr} + \frac{MF \cdot (\Psi_{SR} - \Psi_{cr}) + RSP}{AC}, \quad (\text{A.13})$$

and

$$c = \Psi_{cr} \cdot \left\{ (\Psi_R - z_T) - \frac{RSP}{AC} \right\} - \frac{MF \cdot (\Psi_R - z_T) \cdot (\Psi_{SR} - \Psi_{cr})}{AC}. \quad (\text{A.14})$$

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## Studies on the impact of global climate change on some environmental factors in Hungary

Á. Kertész<sup>1</sup>, D. Lóczy<sup>2</sup>, J. Mika<sup>3</sup>, S. Papp<sup>4</sup>, T. Huszár<sup>1</sup> and A. Sántha<sup>5</sup>

<sup>1</sup>Department of Physical Geography, Geographical Research Institute,  
Hungarian Academy of Sciences, H-1061 Budapest, Andrásy út 62, Hungary;

E-mail: kertesza@helka.iif.hu; E-mail: thuszar@helka.iif.hu

<sup>2</sup>Department of Physical Geography, Janus Pannonius University,  
H-7624 Pécs, Ifjúság útja 6, Hungary; E-mail: loczyd@ttk.jpte.hu

<sup>3</sup>Hungarian Meteorological Service,

H-1024 Budapest, Kitaibel Pál u. 1, Hungary; E-mail: mika@met.hu

<sup>4</sup>Department of Physical Geography, Eötvös Loránd University,  
H-1083 Budapest, Ludovika tér 2, Hungary; E-mail: papps@ludens.elte.hu

<sup>5</sup>Bureau of Natural Protection, Authority for Nature Conservation, Ministry of Environment,  
H-1121 Budapest, Költő u. 21, Hungary; santha.antal@ktmdom2.ktm.hu

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**Abstract**—Within the framework of the MEDALUS (Mediterranean Desertification and Land Use) Project funded by the European Union, authors investigate the impacts of global climate change on the physical environment of Hungary. The Danube-Tisza Interfluvium was chosen as a test area.

Climate change in our region can be characterized by the term *aridification*. Trends of annual mean temperature and of annual precipitation give evidence of warming and drying during the last few decades. The average warming for the last 110 years is about 1 K per century and the change of precipitation is -0.91 mm per year. Because of the importance of the relationship between global and regional variations, regional scenarios were applied. Temperature and precipitation changes are in accordance with these scenarios.

The Danube-Tisza Interfluvium is one of the most severely affected regions of Hungary as far as the drop of free groundwater levels and the depletion of confined groundwater reserves are concerned. 2 to 4 m drops in the annual mean groundwater level (compared to the average of the 1960s, the period, which most certainly preceded the advent of aridification) are recorded. Soil moisture content also reduced during the first half of 1990s. The water level of ponds subsided as well. The dropping ground water level is, however, influenced by many factors so that it is not only the result of aridification.

Vegetation changes of the test area were investigated in detail. From the distribution of floral elements it is claimed that the continental group predominates but the proportion of the mediterranean group is very high (more than one third).

In four fixed cenological quadrates (represented by various subassociations of perennial *Festucetum vaginatae danubiale* of the Danube-Tisza Interfluvium) classical cenological

survey was supplemented with the study of plant species, which may show quantitative changes probably associated with climate change. Four plant species were selected: *Euphorbia seguierana*, *Artemisia campestris*, *Festuca vaginata* and *Stipa borysthena*. The findings of this survey confirm aridification.

Investigations on soil dynamics revealed that in the most sensitive areas, with the gradual lowering of the water table in alkali ponds and with the complete desiccation of some of them, the direct contact between groundwater and salt-affected soils is interrupted, the solonchak soil dynamics ceases, helophile and hygrophile plant associations disappear.

The soda contents of solonchaks effected by a dry period have leached out from the whole profile. This is the most important indirect evidence for desalinization.

Impacts on soil moisture content were studied by applying the EPIC (Erosion Productivity Impact Calculator) model. The method of geographical analogy was applied, i.e. climate data of Pápa were selected as geographical analogues for the expected changes in the Pécsely basin.

Within the frame of a land capability analysis, environmental conditions were assessed for five major arable corps, taking decreasing precipitation amounts into account. The result was a significantly reduced land capability.

*Key-words:* climate scenarios, physical geography, desertification, land degradation, impacts on soils, soil moisture

## ***1. Introduction***

The ever increasing influence of human society on the physical environment may bring irreversible processes which are going to shape the setting for human life in the 21st century. At first, such processes probably occur in the atmosphere, where local and regional influences are most readily globalised, but — through transfers in the system of the physical environment — soon they are also to appear in less dynamic geospheres. Although the processes and consequences involved in global climate change have not yet been fully disclosed by international research, undeniable signs are described from various geographical environments. In the Mediterranean region, for instance, a desiccation tendency or locally even desertification cannot be questioned. In this region of marginal agricultural potential, land degradation has reached a critical stage.

Lying in the very heart of the Carpathian Basin, and on a low-lying flat terrain, Hungary may have to face severe ramifications. Like in other regions of Europe, the meteorological record for recent years in Hungary shows major deviations from long-term mean values. The transitional climate of the country with the three main (continental, mediterranean, and atlantic) climatic influences may easily be “diverted”, rearranged into a new combination of climatic elements which resembles to the conditions of a neighbouring region (probably the Mediterranean) more closely. This involves a moderate warming and a relatively strong inclination to drought, which tendencies are running parallel with global warming processes, mostly likely connected with increasing greenhouse effect (*IPCC*, 1996).

Climate change is in interaction with geographical space and, consequently, it is within the scope of physical geography. A recent concept of geographical implication, i.e. the nature of response to the modification of climate, also supports this claim. It has been suggested that the change, starting from climate and affecting other environmental subsystems, does not simply cause a shift in geographical zones as it was conceived, for instance, when explaining the recolonisation of land liberated from ice-sheets in post-glacial times. It has been found that plant and animal species show variable adjustments, respond to different environmental thresholds and begin to "return" from their refuge areas at various dates (*Graham and Grimm, 1990*). Thus, instead of shifting of entire communities, diffuse dislocations are the rule.

Recalling that water availability is of major importance under the physical conditions of Hungary, the term *aridification* (*Kertész, 1995*) seems to be the most appropriate to describe ongoing processes. In authors' interpretation aridification means an increasing semiarid character manifested over a longer period in rising annual mean temperature, involving a rise in potential evaporation and in decreasing annual precipitation, leading to water deficit. Eventually, transformation of the physical environment ensues as a whole.

The expected impacts of global warming in the Mediterranean are studied comprehensively by the MEDALUS (Mediterranean Desertification and Land Use) project in international collaboration. Most of the research takes place in European countries since the project is financed by the European Union. Research is aimed at modelling predictable transformations in the physical and social environments and to found strategies for sustainable development. Investigations in Hungary constitute subproject MEDEAST (*Kertész et al. 1997, 1998*).

Main objectives of the Hungarian subproject are (see *Fig. 1*):

- I. Climatological research to trace climate change in Hungary: is aridification justifiable from short- and medium-term climatic time series? What scenarios are valid for Hungary?
- II. Research into the physical processes induced by aridification:
  - (a) Changes in groundwater level — particularly in the most drought-stricken area.
  - (b) The influence of climate change on natural vegetation.
  - (c) Soil moisture dynamics and the impact of groundwater level changes on soil processes.
  - (d) The control of aridification on geomorphological processes, e.g., soil erosion.
- III. Land use adjustment to semiarid conditions: surveying and mapping changes and the investigation of land capability.

In our working hypothesis lowlands will show consequences different from those in mountains. Observations support the assumption that, along with the

amount of atmospheric precipitation, its spatial and temporal patterns (distribution and intensity) also change to a considerable extent. In areas of high relief soil erosion conditions are modified, and more intense showers may remove larger amounts of soil. In lowlands, on the other hand, water deficit presents a major problem. In the present paper results from research on the Danube-Tisza Interfluvium test area are dealt with.

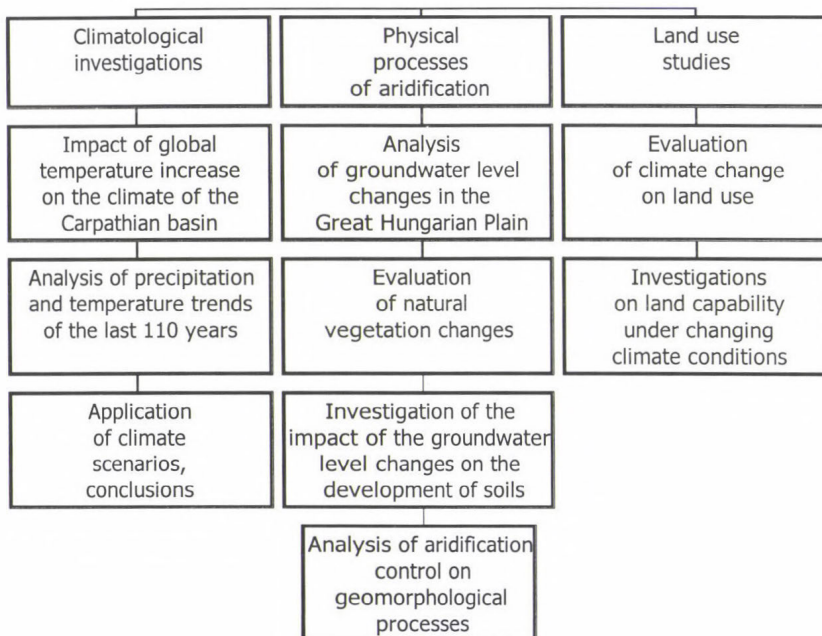


Fig. 1. Aridification research programme.

The response of each factor of the physical environment to global climate change takes place at different rates. The slowest response can probably be expected in the case of geomorphological processes, e.g., in the change of the erosion regime. The rate of vegetation change is rather quick, while soils are also modified relatively fast — although at a slower rate than vegetation. The main driving force in most environments is the reduced availability of water. In arid, semiarid and subhumid environments the concept of desertification is applied to characterize land degradation resulting from various factors, including climatic variations and human activities. Thus the changes of the physical environment described below can also be considered as components of the desertification process.

Taking the time scale of the processes into account, vegetation changes, changes in soil formation processes and in soil moisture will be discussed in detail below preceded by a short analysis of climate tendencies and of groundwater level changes as groundwater table fluctuations are the main driving force of environmental change in the Great Hungarian Plain. Another important aspect, i.e. land capability will shortly be dealt with as well.

## ***2. Evidences for climate and ground water level change***

We briefly overview how the past climate series and climate scenarios (based also on them) support this expectation.

### ***2.1 Observed trends in the meteorological series***

Data series for mathematical trend analysis are available for the past one-or-two centuries only. Systematic observations dating back for more than a century put meteorological data in a very favourable position compared with other environmental data (Nemes and Szalai, 1992). Reference stations to detect climate change of the time series and to represent changes of various climate elements in time and in space have been selected also in Hungary (Ambrózy, 1991).

Time series, however, must be free from inhomogeneities due either to local environmental effects or to the changes of measuring devices. Inhomogeneity means that data series characterising climate variations at the macroclimatic scale were influenced by mezo- or micro-climatic modifications around the site of observation or simply the place, instrumentation, or time of observations, etc. (e.g. Heino, 1994).

In Hungary the measurement frequency of temperature, air moisture, wind direction and wind velocity changed in the middle of the 1960-s which caused an error. This error could be corrected when calculating averages, but recent homogeneity analyses have indicated that the correction made for the climate data series was inappropriate, at least in the case of temperature (Szentimrey, 1994, 1997; Szentimrey *et al.*, 1998).

In the first study, Szentimrey (1994) considered temperature data of a meteorological station (Kremsmünster, Austria) as undistorted references in the mathematical breakpoint detection. Due to this homogeneity correction, the signals became identical at every station of Hungary.

Since *temperature* series have substantial inhomogeneities and the process of correction initiated at the Hungarian Meteorological Service is still in process, temperature analyses will not be dealt with here. According to the first experiences of homogenisation, an increasing trend (cca. 1 K/century) of the annual mean temperature can be observed (Molnár, 1995).

The warming tendency was clearly indicated by Nemes (1994), using yearly absolute minimum temperature values measured at any station of the country,

as this parameter is being measured by different instruments (preserving the minimum measured value).

*Precipitation* series may have inhomogeneities because of changes in the instrumentation, too, but they mainly concern the data prior to 1901 (*Schirok-Kriston*, 1994). One of the mathematically best established calculation (*Koflano-vits-Adámy* and *Szentimrey*, 1986) refers to seasonal precipitation data of 32 stations in the Carpathian basin between 1901–1984. The main results of this study are the maps of linear trends which are negative in spring and autumn, but non-significant (not even at the 90% level) for the rest of the year. Annual precipitation trends are negative everywhere, where significant.

*Ambrózy et al.* (1990) demonstrates how the standard 30-year averages varied between 1901 and 1988 at 162 precipitation stations in Hungary. The conclusion is that even the 30 years' averages varied substantially at a ten year time scale.

Half-year totals of winter precipitation after 1901 were approximated (*Nemes*, 1994) by a quadratic function with a maximum value in the thirties and decreasing afterwards. Ninety year trends of the summer half-year show a significant linear decrease.

17 stations between 1901–1989 were examined by *Molnár* (1995). The stations are mainly located in lowland areas, with the exception of Eger. The mean annual trend for the 17 stations was  $-0,9$  mm/year.

The comparison of *precipitation* averages for the same 17 stations of the two periods (1900–1949 and 1950–1989; *Molnár*, 1995) demonstrates that there is a decrease in most of the months. In annual total, all stations show a decrease of precipitation in the second period (in some parts of the Great Hungarian Plain 10–15 mm, while in other parts 40–60 mm). As for the annual differences of precipitation changes, it is remarkable that there is no precipitation decrease in the summer. A strong increase is observed for June in the second period. Precipitation decline is very remarkable in the autumn months of the second period.

A somewhat different result was gained by *Schirok-Kriston* (1994) concerning precipitations exceeding the 30 mm/day threshold. The analysis of precipitation data between 1901–1990 only for the days above the threshold value does not show a significant trend of increase for Hungary. Precipitation frequency above the threshold was somewhat higher between 1931–60 than for 1901–1930, or for 1961–1990. From among the 30 years' averages prior to 1990, the highest mean temperature of the Northern Hemisphere (e.g. *Jones*, 1994) was reached in 1931–1960, which period was slightly warmer than the period of 1961–1990. This fact draws our attention to the relationship between global and regional variations. This approach is the basis of regional climate scenarios described in the next section. Principal problems of regional climate scenarios and some alternative approaches are dealt with in detail by *Mika* (1993).

## 2.2 Regional climate scenarios

The analysis of small (<1 K) changes of the hemispherical mean temperature was based on time series of instrumental measurements. A method of “slices” was introduced (Mika, 1988) to investigate the connections between regional climatic elements and two hemispherical temperature characteristics, i.e. average temperature (<T>) and air temperature contrast between continents and oceans (DT). The latter characteristic may be close to the optimum, as the first EOFs of CO<sub>2</sub>-forced temperature change patterns (Cubasch *et al.*, 1992) show remarkable coincidence with the continent-ocean distribution map. Hemispherical mean temperature and continent-ocean contrast are derived from air temperatures above the continents by linear weighting according to the areas of the two domains. Correlation of the two hemispherical variables is negligible for all applied divisions of the investigated period, which makes it possible to avoid the problems of multi-collinearity of the physically plausible variables.

The essential point of the method is to divide the original time series into sub-periods of uniform length, and to perform a regression analysis between the time averages for the sub-periods. Namely

$$Y = b_0 + b_1 \langle T \rangle + b_2 DT + e, \quad (1)$$

where  $Y$  is the mean value of the local variable;  $b_i$  is the regression parameters;  $i = 0, 1, 2$ ;  $\langle T \rangle$  is the average of the hemispherical mean temperature for the investigated period;  $DT$  is the average value of the hemispherical ocean-continents contrasts for the same period;  $e$  is the variable with normal distribution.

The aim of “slicing” is to quantify the non-significant relationships of the year-by-year resolution and to randomize the possible inhomogeneities of the series. In addition to the six (5, 9, 13, 17, 21 and 25 years long) sets of time-slices, a set of 13 years long “quasi-equilibrium” slices were defined with negligible air-temperature trends above the continents for the period 1881–1980.

With the method of slices it could be established that the 0.5 K hemispherical warming was accompanied with a 0.5–0.8 K temperature increase, a 7–14 percent decrease of precipitation and with a 20 percent increase in sunshine duration in the summer half-year during the last 100 years. There is a relationship between temperature and the continent-ocean temperature contrast in the winter half-year whereas precipitation and sunshine duration changes could be shown ambiguously. These changes correspond to 5–10 days increase in the vegetation period, about 60% increase in the frequency of dry months (with a soil humidity of less than 30% of available potential) and an 8–10% increase in global radiation at the surface.

The method of slices was also applied for temperature fields of the temperate Northern Hemisphere (Mika, 1992). In these computations,

temperature in Greenland exhibits large relative sensitivity to hemispherical mean temperature variations. This fact was employed to prove the more general character of the relationships for different local variables in Hungary, using the measurements of the independent period 1981–1995, historical data from the period 1490–1779 and paleoclimatic reconstruction from 6000 to 1000 years BP.

Results of larger ( $> 1$  K) global changes are described by *Mika* (1991). The verification of the above scenarios for recent independent periods will only be given here. The “slicing” method was applied for the 100 year period preceding 1980. The following 15 year period can be used as an independent period to validate the scenarios. For this purpose, 28 stations for temperature differences, with no inhomogeneities caused by the changes in the observation times and more than 300 stations for the precipitation differences were employed to determine the local anomalies in the later periods as compared to the 1951–1980 “baseline” climate.

Temperature and precipitation anomalies (*Figs. 2, 3*) in two independent periods after 1981 (1981–90 and 1991–95) broadly correspond to our expectations. This means that the anomalies have the same sign and order of magnitude as the product of the regression coefficients and hemispherical temperature anomalies of the recent periods (i.e.  $+0.21$  K and  $\sim 0.35$  K for  $\langle T \rangle$ ;  $0.16$  K and  $> 0$  for  $\Delta T$ ).

Winter half-year temperatures correspond to the scenario over most of the country (*Fig. 2*). The only exception is in East Hungary with a negative difference between the recent 10 and the preceding 30 years. In the summer half-year (*Fig. 2*) there are positive anomalies in the whole country for the last 10 years.

Precipitation changes are also in accordance with the scenarios (*Fig. 3*). Winter half-year precipitations decreased with the exception of a few small areas in western Hungary. In the summer half-year the strong decrease of precipitation is repeated, except in a few spots (*Fig. 3*). The largest drops are almost 100 mm (i.e., 25–30 per cent of the mean value of the previous 30 years).

It can be concluded that there are local anomalies for the 1981–1990 period corresponding to the scenarios based on statistical relationships from the previous century.

Since early 20th century, a number of evidence have been collected to support warming: winter half-year mean temperatures are rising, summers are ever hotter and the length of the growing season is expanding. Aridification is first of all manifested in the lower number of days with precipitation; rainfall intensity and, as a consequence, runoff probably increases; infiltration and groundwater table are reduced; soil moisture contents (particularly the moisture stored in the soil at the beginning of the growing season) drop and liability to drought grows. For three of the stations the autumn precipitation maximum seems to disappear entirely — in the very regions with marked mediterranean climatic influence.

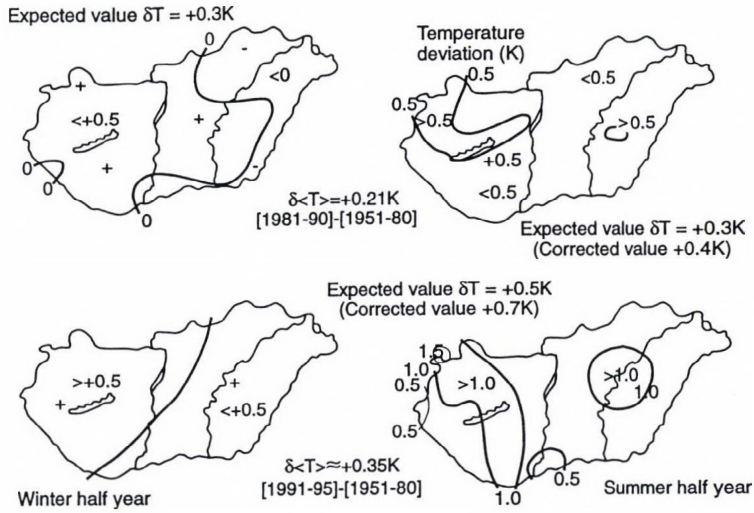


Fig. 2. Temperature deviations of the recent ten year (1981–1990) and five year (1991–1995) periods from the climate normal values of 1951–1980 in the winter half-year (October–March) and in the summer half-year (April–September).

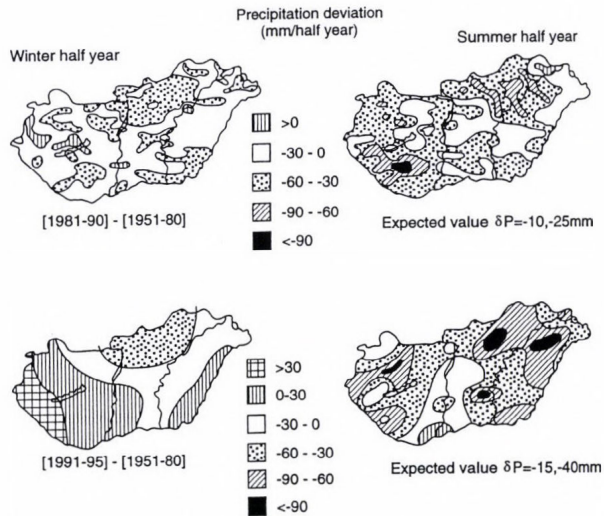


Fig. 3. Precipitation deviations of the recent ten year (1981–1990) and five year (1991–1995) periods from the climate normal values of 1951–1980 in the winter half-year (October–March) and in the summer half-year (April–September).

### 2.3 Groundwater level changes

The Danube-Tisza Interfluve (with a predominant sand soils) is one of the most seriously affected areas by droughts. Here the subsidence of groundwater table has been accompanying climatic tendencies since the 1970s embedded in the above outlined chain of processes. The most serious aspects of aridification here is the extremely reduced infiltration into the soils and the recharge of groundwater (Szász, 1987; Huszár *et al.*, 1996). In 1985–86 groundwater reached its lowermost recorded level (Pálfai, 1991, 1995). A lasting period of drought began in 1983 and since then the frequency of drought years is unprecedentedly high. In the past 15 years cumulative water deficit has amounted to more than 1000 mm. In the humid 1960s depths to groundwater table were recorded at 2 m below the surface with a seasonal range of 0.5 m, and most of the crops could be supplied with sufficient amounts of water (Szalay *et al.*, 1995). The recent shrinkage of free and confined (Berényi and Erdélyi, 1990) groundwater reserves, however, is endangering profitable cultivation in the region (Varga-Haszonits, 1988; Major *et al.* 1991).

As it was demonstrated, the first and most direct impact of aridification is found in the subsidence of the groundwater table. With dropping groundwater, *soil moisture* contents have been also reduced considerably during the 1990s (Pálfai, 1996). For instance, in the spring of 1990 in some sections of the Danube-Tisza Interfluve, the uppermost 1 m of soil had only 60 to 70 per cent soil-moisture reserves as opposed to the long-term average of 100 per cent of field capacity. In 1992 in the same area the 0 to 0.5 m topsoil contained less than 15 per cent moisture, which is below the wilting-point of most agricultural crops. Before the wet winter of 1995, the winter precipitation deficit had maintained a decreasing trend of relative moisture content in topsoil for 12–15 years.

The drought also involves water level drops of ponds. Confined groundwater reserves suffer from increased water intake for irrigation. Their level has sunk recently more than 20 m locally (Berényi and Erdélyi, 1990). The area affected is virtually the same as in the case of free groundwater. After the depletion of the Quaternary aquifer of the alluvial fan, the Pliocene aquifers follow and their pressure conditions are also on the decline.

It is debated whether global climate change is a major and sole driving force of dropping groundwater table on the Danube-Tisza Interfluve. Several authors sound opinions that human activities play an equally great part in the process (Major and Neppel, 1988; Liebe, 1993; Pálfai, 1996). It is well known that afforestation and deforestation influence local water budget. The percentage of the forested area on the Great Hungarian Plain has undergone an important change, i.e. from 25–30% around 1000 a.D. to 6% at the beginning of the 19th century (Bartha, 1993) and rising again to cca. 10% today. About two-thirds of the Great Hungarian Plain were covered constantly by water (swamp, or water surface) preceding drainage works of the 19th century. Drainage reduced

the extension of seasonally inundated areas and swamps, and the water coming from the mountains was transported directly and exclusively by the rivers (Nováky, 1993). These changes induced by human activity during the last 150 years also contributed to increasing aridity of the Great Hungarian Plain. Human influence has been continuously very important since the drainage measures and thus it is difficult and complicated to separate the pure influence of global climate change from human impact. Anthropogenic influence does not only mean water regulation and land use change but it is also manifested in direct water intake, irrigation, amelioration measures and sewage production. Although water intake from confined groundwater for drinking and irrigation is growing steadily, the final cause of increased demand can be detected in the deficit of atmospheric precipitation and increased evaporation (driven by a 3–5 per cent growth in the mean temperature of the growing season). To the joint influence of these factors, groundwater levels dropped over a vast area (almost 9000 km<sup>2</sup>). Although fluctuations of the groundwater level on the Danube-Tisza Interfluvium have ever been observed during the last 100 years, groundwater level subsidence since the 1970s is extremely strong and it can be taken for a long-term trend connected with the above-mentioned human interventions and with global climate change. In the most susceptible areas, first of all in sand regions of the Danube-Tisza Interfluvium 2–4 m drops of the mean annual groundwater level (compared to the average of the 1960s, the period preceding the beginning of aridification) are common (see Fig. 4). The subsidence of the groundwater level may locally reach 5–6 m.

A further depletion of groundwater resources on the Danube-Tisza Interfluvium is expected in the wake of climate change. The shortage of soil moisture and groundwater is the main driving force of aridification in the region leading to the changes of the physical environment described below.

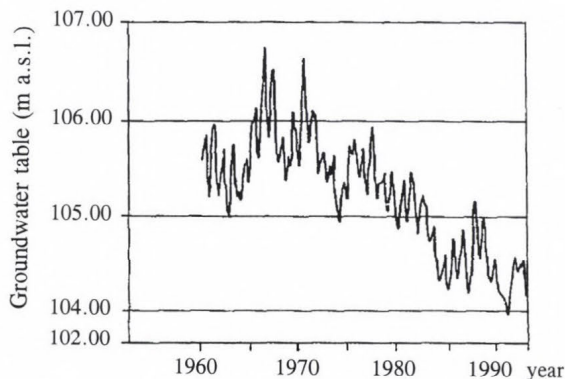


Fig. 4. Lowering of groundwater table (1952–1992) in the well of Ágasegyháza (Great Hungarian Plain)

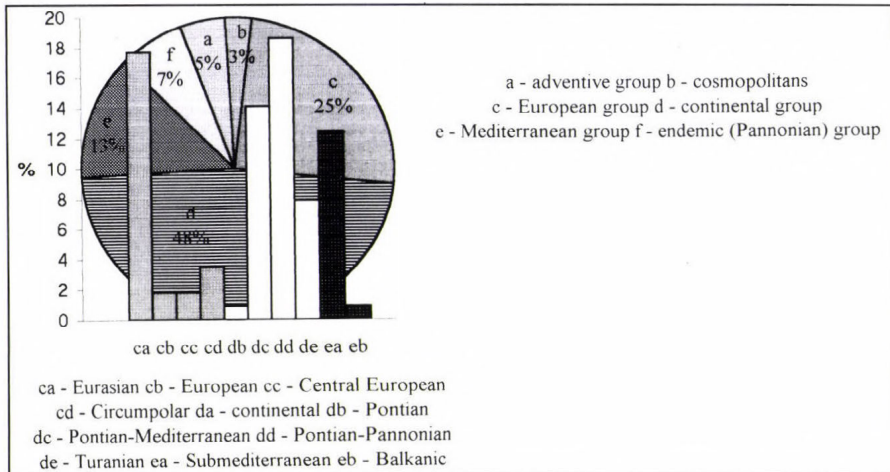
### 3. Changes in the physical environment

#### 3.1 Vegetation changes

Unfavourable hydrological changes induced by human intervention on the Danube-Tisza sand ridge were investigated in earlier research projects of the 1970s and 80s. Naturally, no mention of climate change was made at that time. With regard to the rather recent interest in this topic, research in Hungary including field botanical surveys, have not yet produced significant results. Very often the major task researchers are dealing with is the selection of appropriate methods of investigation and checking their applicability. Even in case of elaborating and employing the suitable methods — regarding the characteristics of the subject studied — findings to be evaluated and interpreted are only expected to be obtained in several years as far as floristic and vegetation surveys directed at aridification are concerned.

It is well-known that climate change is a major control of species distribution and abundance as well as the establishment of living communities (associations). Since naturally induced climate change is a very slow process and each species has a tolerance within broader or narrower limits, there is a certain time lag observed in the response of biota to climate change. Within a longer climatic period, short spells of climatic fluctuation are sometimes detected and their influence on biota is negligible.

According to the recent conception of Hungarian flora, the floral elements in the study area are shown on *Fig. 5*.



*Fig. 5.* Distribution of floral elements of the study area.

From the distribution of floral elements it is claimed that in the study area species of the continental group predominate, species of the European group are half of that number and somewhat more than one third belongs to the Mediterranean group. The ratio of Pannonian (endemic) species is relatively high, while adventives and cosmopolitans are not considerable. Mediterranean climatic influence is primarily characteristic in South-Transdanubia and along the south-central part of the Transdanubian Mountain Range. With regard to this, the expressly high frequency of Mediterranean species is of particular importance. In addition, if we consider the Pontian-Mediterranean elements classed with the continental group, the Mediterranean character is even more pronounced.

In the fixed cenological quadrates (represented by various subassociations of perennial *Festucetum vaginatae danubiale* of the Danube-Tisza Interfluve) classical cenological survey was supplemented with the study of plant species, which may show quantitative changes of specimen number probably associated with climate change, i.e. aridification of Hungarian climate. When identifying the sampling squares, a fundamental condition was the independence of the investigated vegetation type from hydromorphic influence.

Out of the selected four plant species two were dicotyledonous (*Euphorbia seguierana* and *Artemisia campestris*) and two are monocotyledonous graminaceous species (*Festuca vaginata* and *Stipa borysthena*). The graminaceous species are association-forming, prevailing plants in the sand puszta grasslands.

Within the group of Borhidi's relative ecological indicators (Borhidi, 1995) according to the relative groundwater and soil moisture indicator (WB values) *Festuca vaginata*, *Stipa borysthena* and *Euphorbia seguierana* are indicators of habitats with long drought periods (WB = 2), while *Artemisia campestris* represents xerophytes occasionally appearing at moist sites (WB = 3). Thus, the detection of specimen number changes of these four plants in sampling squares can be regarded a simple but effective method of providing evidence for climate change. Botanical observations in the field in recent years have supported the statement that in perennial open sand puszta grasslands *Stipa borysthena* shows an increasing dominance in comparison to *Festuca vaginata*. There is an indirect relationship with the presumed aridification of the Carpathian Basin: the *Stipa borysthena* has deeper root system reaching moister soil layers thus it better tolerates climatic aridification than *Festuca vaginata* with shallower rootage which finds support only in the topsoil.

Considering the above, the joint study of the mass conditions of the four plant species has to be accompanied by the relative quantitative changes of the species, with special regard to the *Stipa/Festuca* ratio.

The *Stipa/Festuca* mass ratio — probably related to aridification — has been investigated in sampling quadrats of summit position, i.e. independently of the microclimatic modification due to exposure. The data of specimen number and coverage point to an overwhelming dominance of *Stipa*: the number of specimens more than fourfold exceeds that of *Festuca*. In other sampling

quadrats of the study area, where the reflecting mass ratios are typical of open perennial sand puszta swards, the ratio of the two species is just the opposite. The finding of this investigation seems to confirm aridification.

Last year the specimen numbers of the four plant species were surveyed from the ten quadrates of 25 m<sup>2</sup> area identified in five different microhabitats of the Fülöpháza test area (Fig. 6). Re-survey is performed every year.

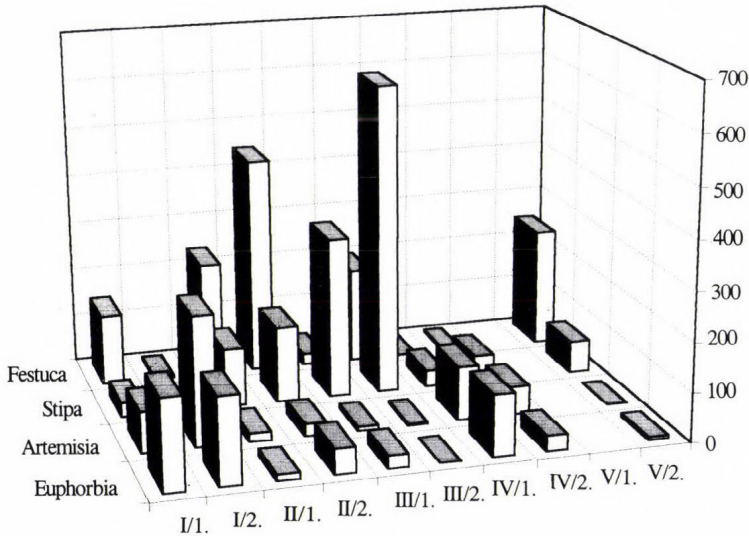


Fig. 6. Distribution of the individuals of the plant species investigated in detail (*Festuca vaginata*, *Stipa borysthenica*, *Artemisia campestris*, *Euphorbia seguriana*) in the quadrates.

It has to be remarked, however, that in order to arrive at interpretable results from the botanical data series, long-term monitoring has to be achieved. This is supported by the properties of Hungarian climate. It suffices here to remind of the interruption of a dry period of 15 years by two humid years and it cannot be judged whether it continues in future or remains a short episode in the process of aridification accelerated by human intervention.

### 3.2 Soil formation processes

Climate change is mediated to the *soil subsystem* through the subsidence of groundwater table and reduction of soil moisture content (Várallyay, 1994). Alterations in water budget are reflected in soil formation with some decades of delay. The statement also applies to the sand soils over the topographic elevations of the Danube-Tisza Interfluve (dune summits and slopes) with

extreme water budget in the first place. Solonchak soils, however, are exceptions and show an extraordinarily rapid seasonal dynamics manifested in the rearrangement of water-soluble potassium salts within the profile.

The investigation of the solonchak-type deposits of partially or totally desiccated alkali ponds is a particularly favorable opportunity to detect land degradation and aridification induced by natural trends (climate change) and human impacts (drainage, water intake, irrigation etc.). Communication between lowered groundwater table and capillary soil moisture is interrupted, soil dynamics towards solonchak formation ceases or even occasional leaching (desalinisation) occurs (*Kertész et al.*, 1998). As a consequence, a type of vegetation different from the previous one — more closed and poorer in halophytic species — can be established.

For the above outlined goals, monitoring has been extended to the immediate neighbourhood of the Danube-Tisza blown-sand area, the dry basin of the Szappan-szék pond. Its study was judged to supplement favorably the investigations into plant succession intended to detect the transformation of the sand region (see previous chapter).

Work began in autumn 1997 with on-the-spot and laboratory analyses of a base soil profile deepened in the most characteristic site of the Szappan-szék. Paradoxically, the soil pit left open the previous year was filled with rainwater during the humid spring and summer of 1998 and thus no sampling could take place. In order to record at least salt dynamics of the first year, samples were taken from an auger hole next to the pit. Both sampling had been preceded by a minimum one-week precipitation-free period.

Results of laboratory analyses of the 1997 and 1998 sampling experiments are presented in *Table 1* and the observed changes of soil properties (total salt and soda contents) are shown on *Fig. 7*.

*Table 1.* Laboratory analyses of base soli profile at Szappan-szék 1. Results indicate changes in the most mobile soil parameters

Depth cm	CaCO <sub>3</sub> %	Humus %	Total salts %		Soda %		pH (H <sub>2</sub> O)	
			1997	1998	1997	1998	1997	1998
0– 18	17.7	2.37	1.10	1.42	0.085	0.446	9.6	10.4
18– 38	14.2	0.21	0.90	0.33	0.080	0.166	9.6	10.2
38– 52	12.1	0.0	1.00	0.20	0.053	0.164	9.5	10.1
52– 71	10.8	0.0	0.80	0.19	0.048	0.144	9.4	10.1
71– 89	15.1	0.0	1.00	0.30	0.053	0.194	9.6	10.2
89–118	33.6	0.0	1.20	*	0.064	*	9.7	*

\* Presence of groundwater made sampling impossible

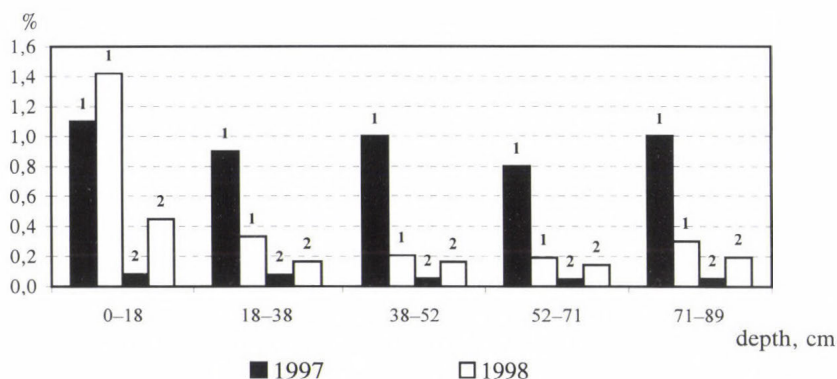


Fig. 7. Changes of the total salt (1) and sodium carbonate (2) content between 1997–1998. in base soil profile of Szappan-szék

The selected profile well represents solonchaks in Hungary with sodium salt content, developed under the influence of high-level groundwater table (obscure horizons, high  $\text{CaCO}_3$  content, strongly alkalic soil reaction [ $\text{pH} > 9$ ]), and — occurring on pond floor — shows more remarkable properties: salt contents including soda exceeds the limits established for this soil type in every horizon and at both measurement dates.

As conclusions it is claimed that:

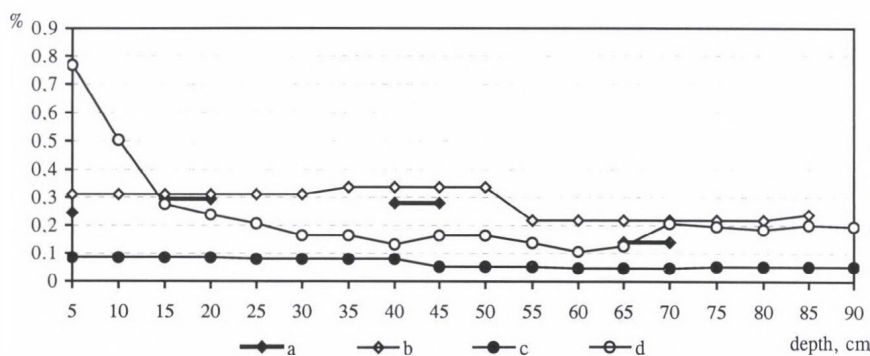
- (1) total salt contents of the soils — with the exception of the uppermost soil horizon — have reduced to one-third or one-fifth;
- (2) soda contents have increased in each horizon and pH grew in parallel, by more than 0.5 (in the uppermost layer by almost 1);
- (3) there is a gradual decrease in the above soil parameters to the depth of 70 cm and reduction is quite abrupt from the uppermost to the next horizon.

Among the above statements, a ready explanation is found for number 2: growth in soda content necessarily involves an increase in pH through alkalic hydrolysis.

Statements (1) and (3) may be related to the more abundant precipitation of the past year and, consequently, to the higher groundwater tables. (In September 1997 groundwater was recorded at 115 cm, while at 72 cm a year later.) In both cases the distribution of salts mentioned is a consequence of differences in salt re-solution and precipitating impacts between the higher groundwater table and the capillary zone above it. With high groundwater levels, most of the salts are dissolved and total salt amounts are reduced. The most easily soluble soda, which remains in solution the longest, was lifted into

the soil profile by capillary processes. The extreme salt content of the uppermost horizon — as it has been shown by the mechanical analysis — is also partly due to the very great share of fine fraction.

A comparison was made between salt contents and salt profile curves of sodaic solonchaks in several dozens of kilometres distance, surveyed in 1980. It was assumed that the comparison may result in recognizing trends of soil dynamics over the almost two decades elapsed. Data from two profiles were described and analyzed by the staff of the Institute of Soil Science and Agrochemistry of the Hungarian Academy of Sciences (*Report*, 1980). The comparison seemed to be suitable (*Fig. 8*).



*Fig. 8.* Spatial and temporal comparison of total soda contents in the solonchak soils of the Danube-Tisza Interfluvium. *a* and *b* = two different profiles described in 1980; *c* and *d* = values of profile at Szappan-szék 1 at two dates (1997 and 1998).

From the analyses of the figures, the following statements can be made:

- (a) Total salt and soda contents are more or less gradually decreasing towards the deeper horizons in both profiles surveyed in 1980; the curves are running close to each other and this indicates similar salt conditions;
- (b) The same parameters of the profile analyzed in 1997 — disregarding minor fluctuations deriving from layering — show similar distribution in the profile (the curves are similar); however, it is striking that curves of total salt and soda indicate much higher or much lower salt amounts than the average (i.e., in profiles described under (a));
- (c) Salt profile curves for 1998 are substantially different from the previous ones: both total salts and soda contents show extremely high values in the uppermost 15–20 cm of the profile.

The following careful conclusions can only be drawn:

- for the time being, from only two profiles, it seems to be a law that salt contents and salt distributions of solonchak soils formed under similar conditions and not yet affected by aridification are largely similar (see (a)); this needs confirmation from the analyses of several further profiles;
- soda contents of solonchaks affected by a dry period of some length (one or two decades) have leached out from the whole profile and equally low in all horizons (see (b)); *this is the most important process and can be indirect evidence for desalinisation;*
- salt conditions developed in dry years can change radically in more humid years when the rising and subsequent lowering of the ground-water table and the reorganisation of the capillary zone — probably temporarily — leads to a concentration of salts in the upper soil horizons again (see (c)).

It is clear for us that measurements have to be performed more often than once a year in the future. Further analyses are necessary (adjusted to seasons, rainfall events, snow melting and considering ion distribution) to explain the processes observed more precisely. As a result, the other objective of research, i.e. to reveal the regularities of plant succession adjusted to salt budget, can also be realised.

### *3.3 Impacts on soil moisture content*

The EPIC (Erosion Productivity Impact Calculator; *Sharpley and Williams, 1990*) is applied to estimate soil water content consequences of the expected regional climate changes in a typical subcatchment, the catchment of Örvényesi-séd, selected for study on the northern catchment of Lake Balaton, according to climate scenarios (*Mika, 1988*). The study is based on the soil hydrology parameters included in EPIC model, exhibiting relatively fast response to the climate variations. To specify regional climate scenarios for Hungary with a coarsely time resolution, two approaches are employed. The simpler one is the use of a geographical analogy which presumes correspondence of differences in monthly variance, and even the daily behaviour of the established differences corresponds to the scenario. Based on this concept, climate data of Pápa (north of Lake Balaton) were selected as geographical analogues for the expected changes in the Pécsely basin, considering the necessary similarities in the non-climatological conditions also. The second approach is a search for statistical connections between semiannual and monthly anomalies, and also between monthly anomalies and daily statistics within the same month. For generating

daily weather sequences, the built-in generator of the EPIC is used. Results of the two approaches are compared. Climate differences generate considerable variation both in the soil water content and in transpiration. Another source of variation is crop-rotation, responsible for the half of standard deviations.

A small partial watershed of 0.3 ha area was selected for comparison. Its average altitude is 202 m, with a slope length of 123 m and a slope inclination of 6 per cent. The slope is covered by a typical, medium-eroded brown forest soil.

### *3.3.1 The EPIC model and its application*

The EPIC was developed to predict the relationship between soil erosion and soil productivity. EPIC is physically based and capable of simultaneously and realistically simulating the processes involved in erosion by using readily available inputs.

Synthetic weather sequences are created by a weather generator. Currently these models are invariably stochastic. The precipitation components of the EPIC weather generator model are first-order two-state Markov chain types (*Richardson and Nicks, 1990*). There are two conceptual subcomponents: an event (or occurrence) submodel and an amount submodel. On each day of the simulation, the event submodel controls whether the day is wet or dry (a wet day is defined as one with at least 0.2 mm of rainfall; *Richardson and Nicks, 1990*). On any day of the simulation a probability for the occurrence of rainfall is chosen randomly. This probability is then compared with a threshold value (the “rainfall state probability”; if it exceeds the threshold then rainfall occurs. The threshold probability depends both on whether rainfall occurred on the previous day (although this is of course unknown for the first day of the simulation), and on the month. Because the occurrence of rainfall on this previous day depends on its turn on the occurrence of rainfall on the day preceding that, and so on, patterns of wet or dry spells extending over several days (“Markov chains”) are built up. Only two states — wet or dry — are stipulated, and the submodel considers only one previous day’s state in determining the threshold probability. It is, therefore, considered to be a first-order, two-state model. The quantity of precipitation on wet days is determined by the amount submodel.

### *3.3.2 Geographical analogy*

Time resolution of the regional climate scenarios is generally insufficient to be directly applied in environmental (hydrological, agricultural, ecological, etc.) impact studies. Hence, down-scaling of the global climate model outputs in space has to be followed by downscaling of the scenarios in time. This task is tried to be solved by application of geographical analogy. This means to find

another site, as a pair to the initial site in question (i.e. Mentshely in our case), where climate at present is similar to the one, expected by the scenario for the given time-slice of the future. The geographical analog is searched with respect to the scenario, described in Section 2.1.

Application of geographical analogy in this case means that diurnal statistical parameters of the meteorological variables at the initial station are introduced into the weather generator of the EPIC as those representing the 'present' climate at that site. Present statistical parameters derived at the geographical analog site are considered as characteristics of the "future" climate at the initial site.

Geographical analogs were derived using the following steps. Seasonal mean climate normal values were calculated for temperature and precipitation series at all stations of the country in the period 1951–1980. For all stations we computed the degree of analogy from the ratios ( $ANT$  = analogy of temperature,  $ANP$  = analogy of precipitation):

$$ANT = (T_{an} - T_{in}) / T_{ch} \quad \text{and} \quad ANP = (P_{an} - P_{in}) / P_{ch}, \quad (2)$$

where  $T_{an}$  and  $P_{an}$  are temperature and precipitation of the (possible) analogue to the corresponding values of the initial station ( $T_{in}$  and  $P_{in}$ ), whereas  $T_{ch}$  and  $P_{ch}$  are the assessed (positive or negative) changes according to the scenario which correspond to 1 K global warming (Mika, 1988). The above values were calculated for the semiannual means (sums), separately.

For the application of geographical analogy and the evaluation of the method, a locality with geographical conditions, similar to those in the Pécsely Basin and meteorological parameters representing future climate had to be found. There were 42 meteorological stations with sufficient data included into this search. Naturally, the time series of the meteorological stations in the Great Hungarian Plain could not be considered since the evolution, structure, drainage and other landscape factors are entirely different in the two regions. After this further limitation, altogether 7 stations could be found to fulfil the above climatological criteria to be a geographical analog. Six of these stations, however, became further rejected due to geographical dissimilarities to the initial site, including erroneous mezo- or micro-scale effects on data quality.

The only selected analog, the area north of Pápa, the border region between the Bakony foothills and the Little Plain, largely corresponds to the geographical conditions of the Pécsely Basin as far as surface structure, landforms and partially soils are concerned. Differences in three main climate elements characterizing these two points for the investigated 1974–85 period, Mentshely (46°58'N, 17°42'E, 332 m a.s.l.) and Pápa (47°23'N, 17°23'E, 131 m a.s.l.) are presented in *Table 2*. Averaging these differences for the summer half-year

i.e., for the growing season of maize in Hungary and also of soil drought, the differences are 0.5 K in temperature and -58 mm (-14% of the value in Menciahely) in precipitation. These differences, and also those for the winter half-year, are broadly consistent with the conditional forecasts concerning a 0.5 K increase of the hemispherical mean temperature, which is about 3–4 decades according to the global forecasts (IPCC, 1996).

Table 2. Average differences (Pápa–Menciahely) in monthly mean values of temperature (T) and precipitation (P), 1974–1985

	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
T (K)	0.7	0.8	0.9	0.6	0.6	0.5	0.6	0.6	0.4	0.3	0.8	0.8
P (mm)	-2	-13	-10	-8	-10	-18	-5	-11	-6	-5	-15	-13

The weather generator of the EPIC model is parameterized both for present (Menciahely) and “future” (Pápa: present) climate according to the 1974–85 period.

At the end of this paragraph we should mention that there is another, more direct way to apply the geographical analogy (Erdős and Mika, 1993). This is the way when the appropriate environmental characteristics are simply compared at the two sites and the difference between them is claimed to be a consequence of climate differences. This direct application requires high similarity of the non-climatic factors or good correspondence of their differences to those likely caused by gradual climate warming.

### 3.3.3 Results of soil moisture simulations

From the wide set of output parameters provided by the model, we selected those exhibiting fast response to the climate differences. The reason for this is that the design of the experiment, simulating present and future time slices, does not allow to simulate the transient modification of slowly varying soil parameters (e.g., mineralization and immobilization, chemical composition, pH, N and P cycling). The selected parameters are soil water content at the root zone and transpiration by the vegetation. The accuracy of EPIC in predictions was evaluated and gave satisfactory results for simulating of the water balance over a long period. Means, standard errors of the mean, ranges, probability distributions of evapotranspiration, runoff and growing season soil water depletion of observed and predicted values were very similar (Steiner *et al.*, 1990). Calibration in our case was restricted to tillage practice and crop

parameters. Water content is calculated by the model according to the artificially generated weather, especially precipitation and thermal conditions of evapotranspiration. Soil moisture is computed in mm, with respect to the water demand of the plant canopy. It is analyzed in annual average and also in August, when generally the annual minimum occurs (*Table 3*).

*Table 3.* Basic statistical characteristics of the simulations with four-year crop rotation

mm	Annual transpiration		Annual soil-water content		Soil-water content in August	
	Average	Standard deviation	Average	Standard deviation	Average	Standard deviation
Pécsely	278	89	49	18	15	53
Pápa	262	90	39	19	12	48

With crop rotation, characteristic of the Pécsely basin, the soil water content exhibits quadriannual fluctuation both in August and in annual average. In more details, enhanced water demand of maize may explain the extremely negative values. Simulations run according to the recent crop rotation, with four year periodicity, transpiration exhibit similar cycles, but with shifted phase. Extreme minimum transpiration occurs in the 3rd year of rotation, which is largely produced by soil conditions, left bare for 3–4 months after harvesting the rape.

Considering these periodicities as disturbances of climatic differences, the simulations are repeated by assuming maize monoculture for the whole generated 30 years period (*Fig. 9*). Water demand of maize is the largest in July-August, that explains the experienced water deficit of 40–50 mm in the root zone. Of course, the expected monoculture for 30 years is an unrealistic assumption in practice. Our aim was only to cancel out the four-year fluctuation, to realise the climatical comparison.

Phase-out of this fluctuation was successful, since standard deviation of the simulated series decreased to its half in the annual soil water content, in its August value and in annual average transpiration, too (*Table 4*). So the difference in the soil water components caused by climatic differences between Pécsely and Pápa became more conspicuous.

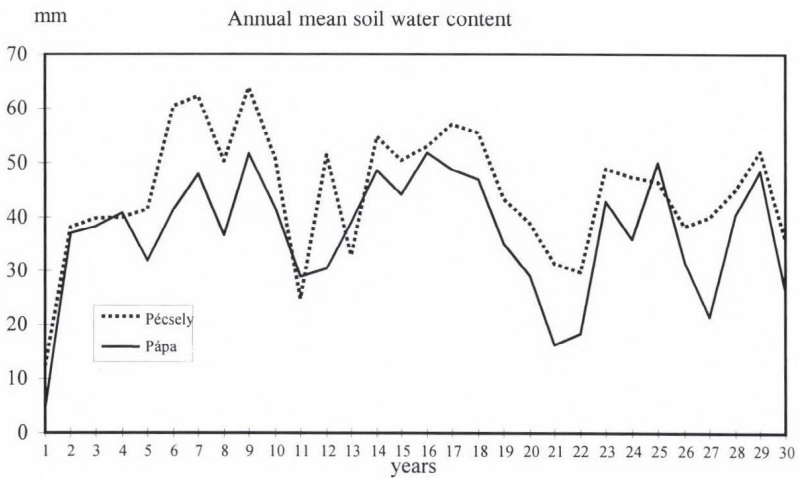
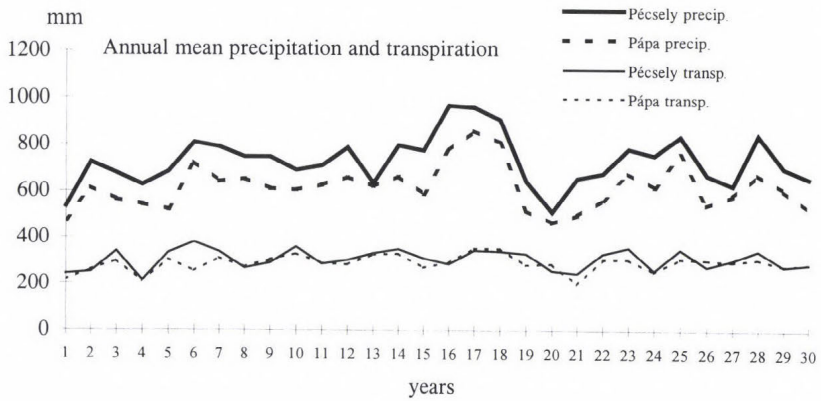


Fig. 9. Simulated annual mean precipitation and transpiration (upper part) and soil water content (lower part) at Pécseley and at its geographical analogue (Pápa) in case of the maize only experiment.

Table 4. Basic statistical characteristics of the simulations with maize monoculture

mm	Annual transpiration		Annual soil-water content		Soil-water content in August	
	Average	Standard deviation	Average	Standard deviation	Average	Standard deviation
Pécseley	306	42	45	12	-36	24
Pápa	289	37	37	11	-43	21

#### 4. Agricultural implications

Through influencing the heat and water balances of landscapes, global warming may significantly affect the physical conditions of farming (Szász, 1987; Hunkár *et al.*, 1995). Even independently from global processes, the climate of Hungary shows liability to summer drought of variable length and the distribution of annual precipitation is irregular (Szalóki, 1994). In Hungary drought is the expression of relative rather than absolute water deficit (Antal, 1991). Drought does not only involve deleterious — occasionally even disastrous — impacts on agriculture when there is not sufficient precipitation but also when no rainwater reaches the root zones of crops (soil drought, or — in soils of poor water storage capacity — when the rainwater does not retained long enough there (physiological drought). Depending on the technology applied, water deficit could result in 10–15 per cent loss of yields.

##### 4.1 Land capability analysis

The deterioration in the conditions of farming caused by reduced water availability have been attempted to be quantified by assessing *land capability* (Lóczy, 1988). As opposed to the usage of the term in Hungarian agricultural literature, land capability here refers to the range of agricultural crops successfully grown in a land area and their order of relative suitability. The agroecological studies in the Geographical Research Institute of the Hungarian Academy of Sciences (Lóczy, 1988; Lóczy and Szalai, 1993, 1994) parameterized water budget by topographic, climatic and soil parameters and portrayed ecological suitability through an ARC/INFO GIS. Environmental conditions were assessed for the five major arable crops (with the largest harvest area): winter wheat, maize, sunflowers, lucerne and sugarbeet. Since the input data incorporated mostly represent the period 1940–1970, the data base can be regarded to reflect conditions before the advent of the aridification trend.

It is assumed that land capability has been reduced by the lack of precipitation, dropping groundwater and lower soil moisture contents since then. In modelling of the changes, attempts to predict the impacts of climate change (e.g., in hydrology: Szilágyi and Vorosmarty, 1993; Kozma-Tóth *et al.*, 1995) have been taken advantage of. The geographical information system for land capability is able to estimate the extent environmental conditions which meet the requirements of crops or groups of crops in the case of dropping groundwater levels and decreasing precipitation amounts. The computer programme of assessment was run with reduced monthly precipitation inputs and evaluated the position of groundwater table relative to the root zones of crops.

## 4.2 Land use

As drought has long been a regular phenomenon over the Interfluve, land use has mostly adjusted to this situation by now. Plantations, primarily the orchards are gaining ground at the expense of arable land. Since the market demand higher is for fruit than for grain, this is an economically favourable tendency. The feedback to groundwater, however, may be positive since orchards — like forests (*Major and Neppel, 1988*) — further reduce groundwater table in their environs.

In general, the impact of forests on water budget needs a careful consideration before judgement. Water demands of arborous species range widely: from black pine (185 mm) to commercial poplar varieties (280 mm). On the long run, losses from the interception of leaves is largely counterbalanced by reduced evapotranspiration under a wetter microclimate. Although forestry experts tend to deny the responsibility of forests in sinking groundwater table, during in areas built up of deposits of low capillarity, forests may contribute to water deficit in neighbouring agricultural land and to reduced agricultural potential drought years without infiltration. Planted apricot and peach trees and vineyards with high summer water demands (*Szász, 1995*) may tap groundwater reserves even more heavily.

## 5. Discussion

Climate change impacts on soil and vegetation are in the focus of the present paper, preceded by a detailed description of climate trends and scenarios concerning Hungary. The main objective is to validate the hypothesis that climate change will cause drier soil conditions in the following decades.

This expectation was shared by various experts (e.g., *HCS D, 1995*). In the recent years there are some doubts about aridification mainly because of the last 3–4 years with normal, or wet conditions. The previous 10–15 years were characterized by dry summer half years corresponding to the warm hemisphere. It is therefore quite possible that 3–4 years ago a new cycle of normal or wet conditions began. We are not yet in the position to answer this question. Further research is needed to clarify whether it is a new cycle or we are facing only 3–4 years with values deviating from the average.

There is a discussion about precipitation change as well. According to several previous GCM outputs, a precipitation increase is projected for Hungary as a response to the CO<sub>2</sub> doubling (see *Mika, 1991* for references). These GCMs were elaborated for 2–4 K temperature increase for which case the statistical methods applied by us were not applicable. This means that there is no contradiction between a drier climate for the 0–1 K global warming and a wetter climate in the case of a 2 K increase.

In a recent experiment performed by the OAGCM at Hadley center, the grid-point data for Hungary (as described by *Harnos*, 1998) gave -14% precipitation decrease for the summer half-year at a CO<sub>2</sub> level corresponding to a 0.7 K global warming. 1.8 K global warming was accompanied by a +8% precipitation increase.

In connection with the scenarios it should also be mentioned that for greater changes there is an *another approach* (*Bartholy et al.*, 1995; *Matyasovszky et al.*, 1995) which combines the frequency of diurnal mid-tropospheric circulation patterns with conditional autocorrelation of the investigated elements (precipitation, temperature and lake-evaporation) to synthesize their future values. These studies applied equilibrium (CO<sub>2</sub>-doubling) atmospheric GCM-outputs only.

The method of geographical analogy, applied in Section 3.3 does not ensure a strictly established method, because modifications of the connections between elements or statistical moments in time could strictly be identical with their variations in space only if we approach climate change by a conservative shift of climate belts (regions) in space. This approach can only be validated if a better method (i.e., the method of slices which works well at longer time scales) could bring good results even at diurnal time scales. According to our first experiences, however, this is not the case. Hence geographical analogy remains a heuristic approach to the downscaling-in-time problem.

## 6. Conclusion

Global climate change is probably manifested in gradual aridification in the SE part of Central Europe. The physical processes triggered, however, are not so advanced as in the Mediterranean region. Aridification tendencies are detected in several environmental factors. The area most affected is the sand region of the Danube-Tisza Interfluve, where aridification — intertwined with human impacts — is expressed in dropping groundwater table, exhaustion of confined groundwater reserves and a slow modification of soil-forming processes. No major change is detectable in natural vegetation and land use.

Research of the consequences of climate change may promote the planning of preparatory measures and reduce environmental and economic damage.

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## Investigation of the chemical composition of bulk precipitation in Tirana, Albania

M. Vasjari<sup>1\*</sup>, A. Cullaj<sup>1</sup> and E. Demiraj<sup>2</sup>

<sup>1</sup>Department of Chemistry, Tirana University, Tirana, Albania;  
E-mail: vasjari@fshn.tirana.al; Fax: +355 42 27625

<sup>2</sup>Institute of Hydrometeorology, Tirana, Albania

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**Abstract**—Precipitation samples were collected in Tirana, Albania during the period June 1995–May 1996, using a bulk precipitation sampler. The daily samples were analyzed for major ions ( $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{NH}_4^+$ ,  $\text{Cl}^-$ ,  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ ), trace metals ( $\text{Pb}^{2+}$ ,  $\text{Cd}^{2+}$ ,  $\text{Zn}^{2+}$ ,  $\text{Cu}^{2+}$ ,  $\text{Fe}^{3+}$ ), conductivity, pH and alkalinity.

The mean pH value of the precipitation, calculated from the volume weighted  $\text{H}^+$  concentration is found to be 6.789. Neutral or alkaline character of rain as a result of neutralization process is primarily caused by calcareous soil dust. Suspended soil contains  $\text{Ca}^{2+}$  ion, which accounts for 68% of the neutralization process. The neutralization factors of  $\text{Ca}^{2+}$ ,  $\text{NH}_4^+$  and  $\text{Mg}^{2+}$  explain their effect in the neutralization process.

In all raining samples  $\text{SO}_4^{2-}$  concentration exceeded the level of concentration of  $\text{NO}_3^-$  and  $\text{Cl}^-$ . If acidity is due to  $\text{nss-SO}_4^{2-}$ ,  $\text{nss-Cl}^-$  and  $\text{NO}_3^-$  concentration,  $\text{nss-SO}_4^{2-}$  was found to be the dominant component during the whole period. The effect of  $\text{NO}_3^-$  and  $\text{nss-Cl}^-$  in acidity of rainwater compensated each other during the period of study.

Contribution of marine sprays to the total concentration of  $\text{Ca}^{2+}$ ,  $\text{SO}_4^{2-}$  was very low. The two fractions were comparable to the level of  $\text{Cl}^-$  only.

The chemical composition data were elaborated taking into account meteorological variables (season of the year, precipitation type, air flow direction) to evaluate temporal variations and chemical source influence. Samples of precipitation arrived from the direction industrialised countries of Central Europe were characterized by the highest concentrations of  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ .

The values of chemical components of Tirana rain samples analysed during this monitoring process were very close to those reported for Thessaloniki, Greece.

*Key-words:* wet precipitation chemistry, major ions, air trajectories.

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\* Corresponding author

## 1. Introduction

Environmental monitoring of precipitation has received particular attention because of the harmful effect of acid rains on the ecosystems and its role as a possible pathway to study the air pollution.

The chemical composition of rain is influenced by both natural and anthropogenic sources. It can not be regarded as a local phenomenon, because it is affected by emissions originated from large regions. Therefore the analysis of chemical composition of rain water connected with meteorological situation can provide a good information on air pollutants as well as on the effects of their long range transmission.

Precipitation chemistry in the Mediterranean area is characterized by a concentration level of main cations ( $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Na}^+$ ,  $\text{K}^+$ ) which is higher than the concentration of main anions ( $\text{Cl}^-$ ,  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ ). It is related to the contribution of carbonate rich aerosols in the air masses travelling from northern Africa to this region. For the same reason, the pH values of precipitation of Southern Europe are higher than in the other parts of Europe. Nevertheless, as the recent studies have been explained, the damaging effects of acid rains were not alleviated even in the case of neutralization (Avila, 1996; Al-Momani, et al., 1995; Samara, et al., 1992). Adriatic Sea is considered to be the most polluted area of the Mediterranean region concerning sulphur compounds, bond nitrogen and heavy metals.

Economic structure of Albania consists of potentially polluting industrial branches as chemical industry, non-ferrous metallurgy, energetic, mechanic and paper industry, etc., emitting nearly all the pollutants reported by the other developed countries. Tirana is the largest city of Albania. The city has grown rapidly since 1991 enforcing too many environmental problems. Particularly the amount of pollutants is increased due to the specific conditions characterized by an uncommonly fast urbanisation. Some chemical and metallurgical industrial plants are situated at North and Southeast direction from Tirana that haven't worked continuously with full capacity during the period of this monitoring process. Tirana is surrounded by Dajti mountain on East. Situated in a field region, about 35 km from the seashore at the East of Adriatic Sea, Tirana is influenced by sea breeze all the year. Also it is influenced by air flow from Northwest to Southeast direction.

In this study one year monitoring data of the rain water of Tirana city are presented. Samples were analysed for  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{NH}_4^+$ ,  $\text{Cl}^-$ ,  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ , trace metals  $\text{Pb}^{2+}$ ,  $\text{Cd}^{2+}$ ,  $\text{Zn}^{2+}$ ,  $\text{Cu}^{2+}$ ,  $\text{Fe}^{3+}$ , conductivity, pH and alkalinity. The relationship between the chemical composition of rain and the meteorological conditions will also be discussed.

## 2. Experimental

### 2.1 Sampling and methods of analyses

Fifty one rain samples were collected during the period June 1995–May 1996, using a bulk precipitation sampler located very close to the centre of Tirana city. Each sample was collected over a 24 or 48 hour period. Samples were filtered through Whatman 41 filter paper and filtrates were subdivided into aliquots and properly conserved accordingly *APHA* (1985) recommendations. After each sampling event the collecting funnel was rinsed with distilled water. It was protected from the dry deposition during dry weather conditions.

Rain samples were analysed by standard methods (*APHA*, 1985). For pH and alkalinity measurements, a Radelkis OP-208/1 pH-meter with a Rose combination glass electrode and an automatic microburette Radelkis OP-930/1 were used. Conductivity measurements were carried out immediately after sampling with a Hanna instrument model HI 8633. Concentrations of  $\text{NH}_4^+$  and  $\text{Cl}^-$  were determined by ionselective potentiometry using Radelkis ISE.  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$  concentrations were measured by UV-method and turbidimetry method receptively, using Pye-Unucam SP6-550 apparatus.  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Zn}^{2+}$ ,  $\text{Na}^+$ ,  $\text{K}^+$  were determined using a Pye-Unicam SP9 AAS. It does not meet the need for additives to suppress interference in rain samples. Other heavy metals have been determined by Electrothermal AAS using a Varian SPECTRAA 10/20 equipment.

Field blanks for wet deposition samples were collected. Approximately 200 ml of distilled deionized water was poured through the funnel and collected in the collection bottle like a rain sample. Sample handling and analytical procedures applied to rain water blanks were the same as those applied to samples.

### 2.2 Quality assurance program

This monitoring process consists of the following elements:

- (1) Consistency checks (*APHA*, 1985; *Kulshrestha et al.*, 1995)

The consistency checks used the evaluation of ion balance, comparing the sum of anions and cations, and the estimation of conductivity calculated from the concentration of each ion multiplied by the equivalent ion conductivity.

- (i) anion-cation balance

The control is based on the electroneutrality of rain water samples. Theoretically the sum of anions expressed in  $\text{mequ L}^{-1}$  must be equal exactly to the sum of cations, similarly expressed in any samples. A control chart was constructed

to identify if the difference between the sums of the cations and anions falls between the control limits. The majority of samples was found to comply this requirement.

(ii) comparison between measured and calculated conductivity

An alternative way for data checking is the plot of measured versus calculated conductivity (APHA, 1985; Samara *et al.*, 1992). The linear correlation coefficient between them exceeds 0.913 (0.0000;  $n = 32$ ).

(2) Quality control chart

Control performance (C/P) chart were use to ensure that quality of distilled water, pH, conductivity, alkalinity and  $Mg^{2+}$  were accurately determined. A further graphical presentation of the data was given using the modified Youden's plot (Q;D;T) (Miller, 1988; Nadkarni, 1991; Meglen, 1985). Firstly the control samples requested for each parameter were prepared, then control limits were calculated based on the firsts 20 measurements.

(3) Participation in the intercalibration exercises

“Aquacon-Med Bas-Subproject Nr. 6 Acid rain analysis” organised by Italian Institution of Hydrobiology during the period of study were used as an alternative way to evaluate the quality of analytical measurements (AQUACON, 1994; AQUACON, 1995).

### 3. Results and discussion

#### 3.1 Chemical composition of rain

Table 1 presents the average composition of the rain water samples as well as the standard deviation values, minimum and maximum values to show the important annual variability.

The volume weighted mean is the best estimate of the average chemical composition of rain water for an annual period (Samara *et al.*, 1992; Avila, 1996; Gatz *et al.*, 1995a, 1995b; Sequeira and Lung, 1995). It was found to be lower than the arithmetic mean for all parameters. Similar relations have been reported in the other studies on rain water composition (Samara *et al.*, 1992). The values of geometric means for all parameters are lower than arithmetic means. This suggests that the log-normal approach is the best for the frequency distribution of these parameters. “Chi squared “ test was used to check the distribution of all parameters (Massart, 1988). It was found that some parameters show bi-modal distribution (pH, conductivity,  $Ca^{2+}$ ,  $Mg^{2+}$ ,  $SO_4^{2-}$ ,  $NH_4^+$ ), perhaps indicating the presence of two classes of rain events.

Table 1. Chemical characteristics of rain (\*  $\mu\text{eq L}^{-1}$ ; \*\* pb)

Parameter	Mean	V. W. M <sup>+</sup>	G. M. <sup>++</sup>	S. D.	Range	n
pH	6.73	6.78	6.71	0.52	4.21-7.41	50
Alkalinity	0.13	0.13	0.12	0.07	0.04-0.39	48
Conductivity	45.24	41.83	41.66	18.49	17.8-99.4	50
(Na <sup>+</sup> )*	46.96	43.28	37.88	32.25	11-129	51
(K <sup>+</sup> )*	10.52	9.97	6.91	11.63	2-61	50
(Ca <sup>2+</sup> )*	233.78	219.44	212.92	105.44	76-644	51
(Mg <sup>2+</sup> )*	27.76	25.30	24.82	13.36	9-68	51
(NH <sub>4</sub> <sup>+</sup> )*	68.08	76.87	44.15	61.70	5-211	34
(NO <sub>3</sub> <sup>-</sup> )*	48.30	41.66	35.27	41.93	5-179	40
(Cl <sup>-</sup> )*	62.71	55.57	46.93	51.41	12-130	46
(SO <sub>4</sub> <sup>2-</sup> )*	152.71	152.47	141.21	61.24	38-345	46
(Zn <sup>2+</sup> )**	56.07	48.94	47.21	36.44	20-160	51
(Cu <sup>2+</sup> )**	5.68	5.02	3.59	7.58	0.4-43.8	50
(Pb <sup>2+</sup> )**	1.06	0.94	0.71	1.08	0.1-6.4	49
(Cd <sup>2+</sup> )**	0.50	0.39	0.25	0.87	0.1-5.6	48
(Fe <sup>3+</sup> )**	76.40	76.90	60.45	57.84	20-310	47

+ Volume weight mean; ++ Geometric mean; S.D. Standard deviation

Table 2 shows that the means of all parameters of rainwater in Tirana are higher compared to that observed elsewhere in the Mediterranean countries. Particularly the closest values were measured in Greece. High values of Ca<sup>2+</sup> suggest that neutralization is the major factor for the low acidity. The manual sampling methods have been affected by the value of Ca<sup>2+</sup> which can be seen in the blank samples too.

According to this study the neutralization process is primarily due to Ca<sup>2+</sup> concentration followed by NH<sub>4</sub><sup>+</sup> and Mg<sup>2+</sup>. The role of Ca<sup>2+</sup>, NH<sub>4</sub><sup>+</sup> and Mg<sup>2+</sup> has been evaluated by calculating neutralization factors (NF) using the formula (Kulshrestha et al., 1995):

$$NF_x = \frac{X}{NO_3^- + SO_4^{2-}}, \quad (1)$$

where X may be Ca<sup>2+</sup>, Mg<sup>2+</sup>, NH<sub>4</sub><sup>+</sup> in mequ L<sup>-1</sup>.

Table 2. The chemical composition of rain water in some Mediterranean countries ( $\mu\text{eq L}^{-1}$ )

	France <sup>(1)</sup>	Spain <sup>(2)</sup>	Turkey <sup>(3)</sup>	Greece <sup>(4)</sup>	Italy <sup>(5)</sup>	Albania
pH	5.0	-	5.6	5.5	5.8	6.8
Alkalinity	-	13.5	-	-	-	134.0
Conductivity	46	-	-	40.2	-	41.8
Na <sup>+</sup>	36	22.3	117	23.5	424	43.3
K <sup>+</sup>	24	4.0	17	8.2	15	10.0
NH <sub>4</sub> <sup>+</sup>	70	22.8	43	54.6	20	76.9
Ca <sup>+</sup>	101	56.6	81	219.0	154	219.4
Mg <sup>+</sup>	19	9.7	101	21.2	115	25.3
Cl	71	28.5	117	35.6	474	55.6
NO <sub>3</sub> <sup>-</sup>	68	20.6	23	47.9	26	41.7
SO <sub>4</sub> <sup>2-</sup>	128	46.0	66	158.0	103	152.5
Site of sampling	Strasbourg	Monstény	Izmir	Thessaloniki	Sardinia	Tirana
Sampling period	1991-1992	1983-1994	1993	1989-1990	1990-1991	1995-1996
Sampling	wet only	bulk	wet&dry	wet only	wet&dry	manual

(1) Sanusi *et al.*, 1996; (2) Avila, 1996; (3) Al-Momani *et al.*, 1995; (4) Samara *et al.*, 1992; (5) Guerzoni *et al.*, 1995

Fig. 1 shows the neutralization effect of these ions. It seems clearly that the contribution of NH<sub>4</sub><sup>+</sup> has a contrary tendency to contribution tendency of Ca<sup>2+</sup>, meanwhile the effect of Mg<sup>2+</sup> is constant during all the period. This situation means that acidity of samples was buffered on equal level. The values of pH ranged from 6.6 to 7.1 in 72% of the samples.

The concentration of SO<sub>4</sub><sup>2-</sup> was higher than that of NO<sub>3</sub><sup>-</sup> and Cl<sup>-</sup>. A study in Thessaloniki gives similar results for these anions (Samara *et al.*, 1992).

Non sea salt fraction of SO<sub>4</sub><sup>2-</sup> (nss-SO<sub>4</sub><sup>2-</sup>) plays an important role in rain acidity due to the oxidation process of SO<sub>2</sub>. NO<sub>3</sub><sup>-</sup> and nss-Cl<sup>-</sup> have an influence on rain acidity, too. The term non sea salt has been used in preference to non marine fraction. Assuming that all the sodium in rain water is derived from the sea nns-fractions were calculated for anions SO<sub>4</sub><sup>2-</sup> and Cl<sup>-</sup>, as follows:

$$x_i = X_i - C_{i,Na^+} \cdot R_d, \quad (2)$$

where  $x_i$  - nss-fraction of anion in sample  $i$ ,  
 $X_i$  - concentration of anion in sample  $i$ ,  
 $C_{i,Na^+}$  - concentration of  $Na^+$  in sample  $i$ ,  
 $R_d$  - ratio of anion X to  $Na^+$  in sea water.

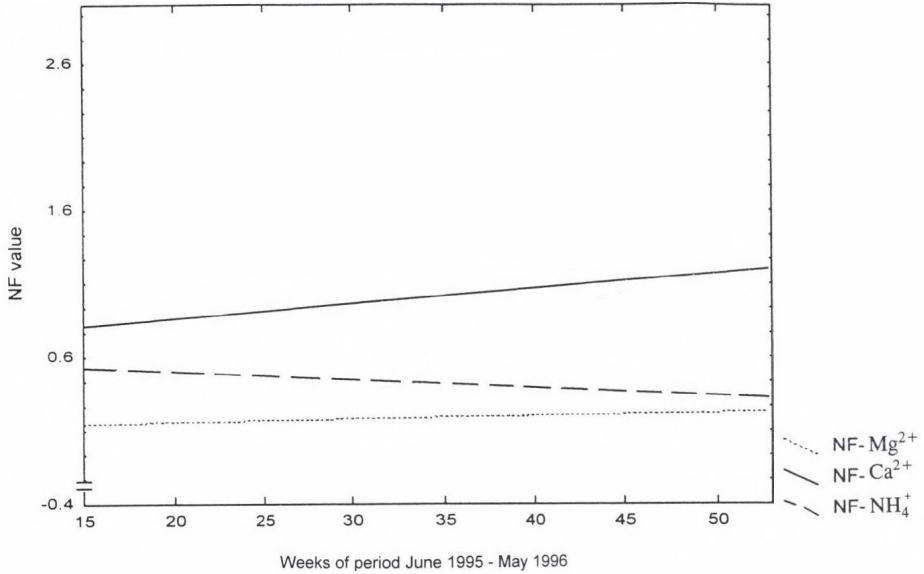


Fig. 1. Neutralization of rain water by  $Ca^{2+}$ ,  $Mg^{2+}$  and  $NH_4^+$ .

Fig. 2 shows the contribution of anions during all the period. It seems that  $SO_4^{2-}$  has dominant role in the acidity of the rain water. The relative contribution of nss- $Cl^-$  and  $NO_3^-$  to the acidity of rain water is variable but they compensate each other. If all the concentrations of nss- $SO_4^{2-}$ , nss- $Cl^-$  and  $NO_3^-$  in our samples were in respective acidic form, this would produce an average pH of 3.7 in the precipitation. Consequently, the high pH observed indicates a neutralization process caused by basic species incorporated in rain water.

The concentration of heavy metals in rain was lower than that of reported for the Mediterranean countries (Amann *et al.*, 1992).

The contribution of each ion to the sum of cation, anion and total ion mass in precipitation is shown on Fig. 3.

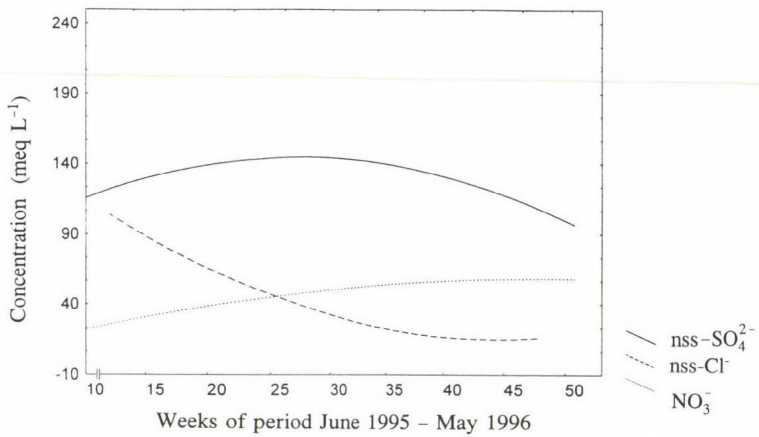


Fig. 2. Contribution of anions to the acidity of rain water.

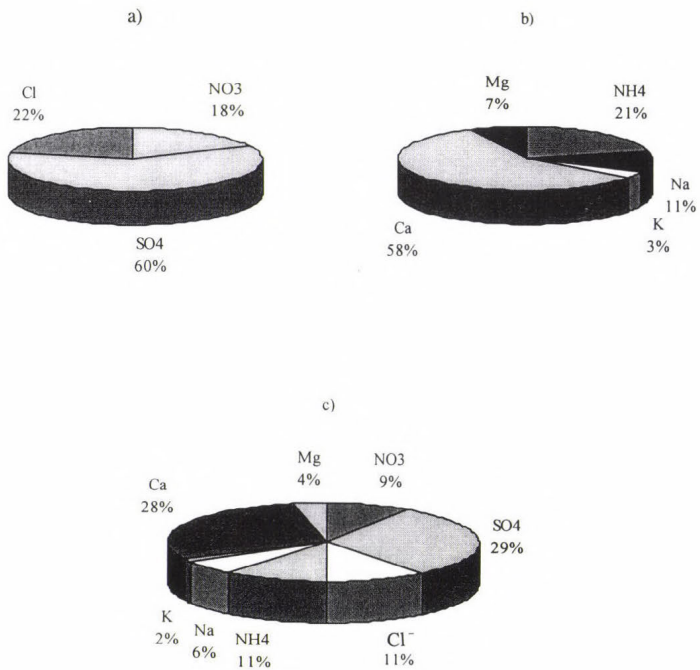


Fig. 3. Contribution of ions to chemical composition of rain water.

### 3.2 Sources of ions

Two methods were used to evaluate the sources of rain pollution. The most usual method of evaluating the contribution of sea salts to ion concentrations in rain water is to compare the ion/Na<sup>-</sup> ratio in rain to that in sea water (*Samara et al.*, 1992). *Table 3* shows the ion/Na<sup>-</sup> concentration ratios and the respective values in sea water.

*Table 3.* Ion/Na<sup>+</sup> concentration ratios in sea water and in our samples

Ion/Na <sup>+</sup> ratio	Ion/Na <sup>+</sup> concentration ratios			
	in sea water	in our samples		
		V.W.M.	G.M.	Mean
K <sup>+</sup> /Na <sup>+</sup>	0.02	0.28	-	0.28
Mg <sup>2+</sup> /Na <sup>+</sup>	0.22	0.72	0.66	0.74
Cl <sup>-</sup> /Na <sup>+</sup>	1.17	1.51	1.27	1.65
Ca <sup>2+</sup> /Na <sup>+</sup>	0.04	6.91	5.63	6.74
SO <sub>4</sub> <sup>2-</sup> /Na <sup>+</sup>	0.12	4.95	3.85	4.60
SO <sub>4</sub> <sup>2-</sup> /NO <sub>3</sub> <sup>-</sup>	-	6.48	3.90	5.76

G.M. Geometric mean; V.W.M. Volume weight mean

Sea is considered to be the only source of sodium while the other ions may also be emitted from other industrial or natural sources. With this assumption we calculated the marine contribution of each ion that is incorporated in marine aerosols.

These contributions are graphically compared on *Fig 4*. It seems clearly that the nss concentrations are higher than the ss concentrations for every ions except Cl<sup>-</sup>. Marine fraction of Cl<sup>-</sup> is higher than non-marine fraction and has considerable value. Meanwhile for the other ions marine contribution is either weak or negligible.

Non-marine fraction of Ca<sup>2+</sup> in rain is about 100 time higher than its marine fraction. A first explanation is the sampling method using a manual bulk precipitation sampler. The main source of atmospheric Ca<sup>2+</sup> is believed to be the soil dust. It could be emitted by human activities such as traffic. In addition air masses originated from North Africa are rich in calcareous soil dust and may bring additional Ca<sup>2+</sup> ions.

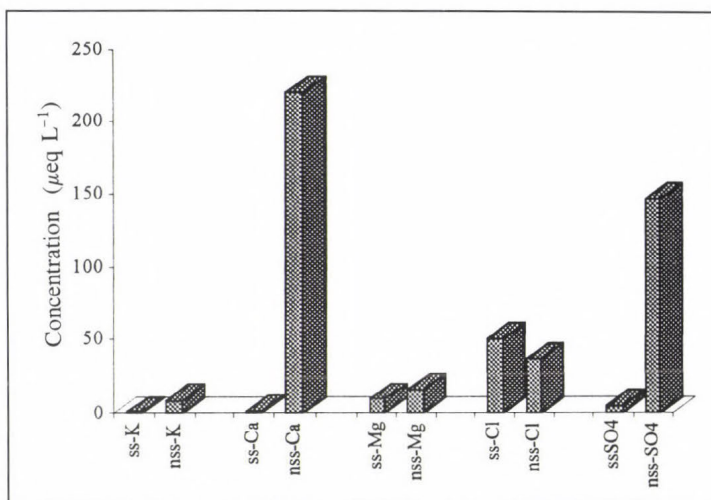


Fig. 4. Sea-salt and non-sea-salt concentrations.

The concentration of  $\text{SO}_4^{2-}$  in precipitation can be attributed to the oxidation process of  $\text{SO}_2$  emitted from the local industry, and to the long-range transport from the industrialized countries.

Correlation analysis was used to obtain additional information on the sources of ions in the rain samples.

$\text{Na}^+$  and  $\text{Cl}^-$  ions are correlated with a coefficient of 0.6309 (44;0.0000) due to their common marine source. Most of the crustal elements, namely  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$  are also correlated with each other with a coefficient of 0.7653 (47;0.0000). Their non-marine fractions ( $\text{nss-Ca}^{2+}$ ,  $\text{nss-Mg}^{2+}$ ) have better correlation (0.8225;43;0.0000). The  $\text{Mg}^{2+}$  ion is also correlated with  $\text{Na}^+$  (0.7346;48;0.0000), indicating that a fraction of this element can be accounted for by sea salt.

Good correlations were found between  $\text{SO}_4^{2-}$  and  $\text{Ca}^{2+}$  (0.6689;44;0.0000), and  $\text{SO}_4^{2-}$  and  $\text{Mg}$  (0.6621;45;0.0000) which support the assumption that solid air particles may be potential sources for  $\text{SO}_4^{2-}$  in precipitation.

As shown from the inspection of the correlation matrix there is no correlation between  $\text{NO}_3^-$  and any other ions. This suggests that  $\text{NO}_3^-$  in rain originates from ionisation of nitrate salts which are produced by neutralization processes.

The quite good correlation of heavy metals ( $\text{Cu}^{2+}$ - $\text{Pb}^{2+}$ ;  $\text{Zn}^{2+}$ - $\text{Cd}^{2+}$ ;  $\text{Cu}^{2+}$ - $\text{Cd}^{2+}$ ;  $\text{Pb}^{2+}$ - $\text{Cd}^{2+}$ ;  $\text{Cu}^{2+}$ - $\text{Zn}^{2+}$ ) with linear correlation coefficients from 0.66 to 0.87 (47;0.0000) indicates their common anthropogenic sources.

### 3.3 Effects of meteorological variables

#### 3.3.1 Seasonal effects

The chemical composition of precipitation varies in different seasons. While the conductivity (VWM) is constant during spring, a seasonal pattern is shown for other measured parameters in summer and autumn.

Concentration of ions that are dominant in chemical composition of rain ( $\text{Ca}^{2+}$ ,  $\text{SO}_4^{2-}$ ) appears to be constant almost through the year without any seasonal tendency. Their concentrations vary from 200 to 400  $\mu\text{eq L}^{-1}$ , and from 100 to 250  $\mu\text{eq L}^{-1}$  for  $\text{Ca}^{2+}$  and  $\text{SO}_4^{2-}$  in most of the rain events. The parameters that show wide range of variability are  $\text{K}^{2+}$ ,  $\text{Cl}^-$  and  $\text{Na}^+$ .

Parameters, which are naturally coupled like pH and alkalinity show the same trend with the highest values in summer.  $\text{Ca}^{2+}$  content also shows its highest values in summer due to the influence of the warm and dry weather. The influence of weather conditions is reflected in contribution of soil dust, too. In calm weather conditions  $\text{Ca}^{2+}$  content exceeds 60.2% and 50.8% in spring and summer, respectively. Thus in spring, when calm weather conditions are more frequent, the contribution of soil dust in the concentration of these cations in the atmosphere of Tirana city is larger.

Ions, primarily originating from marine aerosols ( $\text{Na}^+$ ,  $\text{Cl}^-$ ) appear with lower values during the spring and achieve the highest level of their concentrations in winter. This allows to suggest that during the warm and dry summer the atmosphere is richer in marine aerosols that is reflected in increased value of ion ( $\text{Na}^+$  and  $\text{Cl}^-$ ) concentrations in the following seasons.

*Fig. 5* shows VWM concentrations of  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ ,  $\text{NH}_4^+$  and  $\text{Ca}^{2+}$  ions in March, April and May when fertilizers are mostly used (*Al-Momani et al.*, 1995).  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$  and  $\text{NH}_4^+$  ions have a trend similar to that of the constituents of fertilizers, which suggests that fertilizers play an important role in the composition of rain water in this period. Meantime,  $\text{Ca}^{2+}$  ion shows a different temporal variation as expected.

There were not any significant differences in the seasonal concentrations of heavy metals.

#### 3.3.2 Effects of wind directions

Meteorological variables such as air mass trajectories play an important role in the distribution of air pollution. The air flow direction in the higher level of the atmosphere influences the wind direction in the lower atmospheric layers. Surface winds measured in every hour were taken into account to relate rain composition to rain pollution origin. Frequency distribution of wind direction is given on *Fig. 6*, which shows that the chemical composition of rain during the period of study was strongly affected by NW direction and less strongly by SE, SW wind directions.

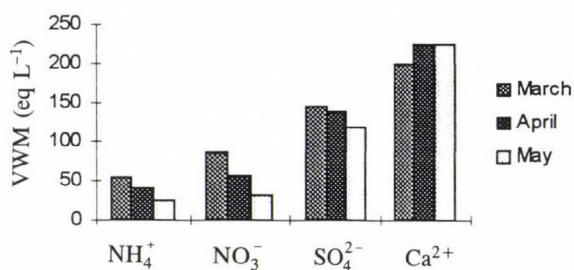


Fig. 5. Role of fertilizers in chemical composition of rain.

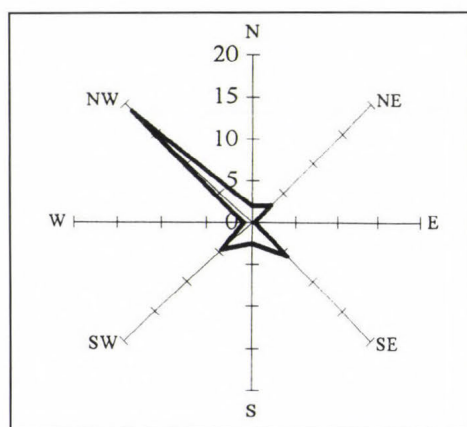


Fig. 6. Wind frequency direction (June 1995–May 1996). Q = 67.3%.

Rain samples are classified into six classes according to wind sectors. Rain samples effected by NW wind direction contained high concentrations of  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$ . It is likely due to the influence of long-range transport of  $\text{SO}_2$  and  $\text{NO}_x$  from industrialised countries of Central Europe. The emissions of local industry are also transported. Their pH, due to the neutralization process extend to the level of the other classes.

Rain samples influenced by SE, SW were characterized by relatively high value of pH. This alkalinity can be attributed to the calcareous soil dust transported from North Africa, or from shorter distances.

The VWM concentrations of  $\text{Na}^+$  and  $\text{Cl}^-$  do not show any trend according to wind direction as it can be expected. This indicates that Tirana region is effected by sea breeze during the whole period investigated.

Rain events are classified into rain and storm. The influence of marine aerosols presented by  $\text{Na}^+$  and  $\text{Cl}^-$  concentrations was the highest in rain cases and the lowest in storm cases, probably due to the fact that most storms did not come from the sea direction.  $\text{NO}_3^-$  showed similar picture. The other parameters did not show significant variations with precipitation type.

Concentration of chemical parameters of Tirana rain samples analysed during this monitoring process were higher than those of reported for some Mediterranean countries, but were very close to those reported for Thessaloniki (Greece).

#### 4. Conclusions

Monitoring of chemical composition of precipitation in Tirana was carried out according to a one year monitoring program based on standard analytical methods and quality assurance programs. The main features of the precipitation chemistry were:

- (1) Rain samples showed neutral pH values, similar to those reported for some other Mediterranean countries.
- (2) Neutralization process was attributed primarily to high concentration of  $\text{Ca}^{2+}$  ion (68%) followed by  $\text{NH}_4^+$  (24%).
- (3) The concentration of  $\text{SO}_4^{2-}$ , was higher than that of nitrates and chlorides.  $\text{Nss-SO}_4^{2-}$  plays the dominant role in acidifying the rain water.
- (4)  $\text{SO}_4^{2-}$  and  $\text{Ca}^{2+}$  ions make the highest contribution to the chemical composition of rain. They account for 29% and 28% of the total mass of ions.
- (5) The concentrations of heavy metals in rain were low, particularly  $\text{Pb}^{2+}$  and  $\text{Cd}^{2+}$  (0.1–7 ppb) were lower than those reported for other Mediterranean areas; whereas the level of  $\text{Zn}^{2+}$  (20–160 ppb) is closer to them.
- (6) Concerning the sources of ions in the chemical composition of rain, the contribution of sea was found to be important on the level of  $\text{Cl}^-$ , whereas for other ions marine effect is negligible.
- (7) During warm and dry summer conditions the atmosphere is mostly influenced by soil dust, and the concentration of crustal elements ( $\text{Ca}^{2+}$ ,  $\text{K}^+$ ,  $\text{Mg}^{2+}$ ) and the parameters that are connected to them (pH, alkalinity) had their highest values.
- (8) The rain samples influenced by the air flow originating from industrialized countries of Central Europe were distinguished by high concentrations of  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$ . At the same time, a good linear

correlation between  $\text{SO}_4^{2-}\text{-Ca}^{2+}$  and  $\text{SO}_4^{2-}\text{-Mg}^{2+}$  could be explained by the influence of solid air particles in the  $\text{SO}_4^{2-}$  concentration in composition of rain water.

- (9) Chemical composition of precipitation varies seasonally. Spring is characterized by more calm weather situations, thus the contribution of soil dust to the concentration of these cations in the atmosphere of Tirana city is larger.
- (10) Chemical composition of the rain water is also affected by the constituents of fertilizers ( $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ ,  $\text{NH}_4^+$ ) particularly during the period of their application.

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