A STUDY ON THE INFLUENCE OF SEDIMENT IN THE PHOSPHOROUS CYCLE IN LAKE BALATON

Peter Gelencser
Ferenc Szilagyi
Laszlo Somlyody
Lambertus Lijklema

July 1982
CP-82--- 44

INTERNATIONAL INSTITUTE FOR APPLIED SYSTEMS ANALYSIS
2361 Laxenburg, Austria

THE AUTHORS

Péter Gelencsér and Ferenc Szilagyi are with the Research Centre for Water Resources Development (VITUKI), Budapest, Hungary.

László Somlyódy is with the International Institute of Applied Systems Analysis, Laxenburg, Austria, on leave from the Research Centre for Water Resources Development (VITUKI), Budapest, Hungary.

Lambertus Lijklema is with the Department of Water Purification, Agricultural University, Wageningen, The Netherlands.

PREFACE

One of the principal projects of the Task on Environmental Quality Control and Management in IIASA's Resources and Environment Area is a case study of eutrophication management for Lake Balaton, Hungary. The case study is a collaborative project involving a number of scientists from several Hungarian institutions and IIASA (for details see WP-80-187 and WP-81-108).

As part of the case study, different lake ecological models and water quality management models are under development. The sediment and its interaction with the water is of major importance for the development of both types of model, since the sediment, as an internal nutrient source, influences the short-term dynamics of water quality and also the long-term changes. Consequently, acceptable knowledge on the behaviour of the sediment is required for the calibration and validation of lake ecological models on one hand, and for the estimation of the new equilibrium of the lake and the renewal time following a load reduction on the other hand. This latter point is a major issue from the point of view of water quality management. This report is intended to serve as a background for handling both types of problem.

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INTRODUCTION

The sediment layer* is one of the key elements in the nutrient cycle in lakes. For most systems, including Lake Balaton, phosphorus is the major nutrient, being the limiting element of eutrophication and perhaps the only one through which feasible management alternatives can be realized for improving the water quality.

In lake Balaton retention is about 95%. The phosphate stored in 1cm of sediment is about two orders of magnitude higher than in the 3 meters of overlying water. Hence the sediments have considerable potential for

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*The concept of sediment or bottom layer is used here to emphasize that the influence of both the sediment and pore water in this layer is considered. Later on the word "layer" has been omitted in cases where it might cause ambiguity.

sustaining biological processes in the water column. The actual rate at which phosphate (via several interrelated processes) is released into the water is a key parameter in the phosphate budget and in the eutrophication process. This release is often called "internal loading". With the development of man-made eutrophication, an increase in the internal load is expected.

From the previous paragraph it follows that the scientific understanding of in-lake processes as a first step and possible management of the system as a second step cannot be properly realized without incorporating the influence of the sediment. In many cases mathematical modeling or, in a broader sense, a systems analytical approach is inevitably required in order to attain the objectives described above, since otherwise the many diverse, though interrelated, processes and their contribution to eutrophication as a whole cannot be adequately tackled. If such a procedure is adapted, as it was for example for Lake Balaton, (see van Straten and Somlyódy 1980, Somlyódy 1981), the sediment should also be included in the modeling framework.

Three important questions should be considered:

(i) What is the magnitude of the internal load compared to the external load, in other words, what is the importance of sediment in a global sense? The external phosphorous load for Lake Balaton is approximately 1000 kg/d (the volume and surface area related values are $0.52 mg / m^3 d$* and $1.68 mg / m^2 d$, respectively) half of this is

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*It should be noted that the volume-related load shows a pronounced longitudinal gradient along the lake (see Figure 3) and it is more than ten times larger at the most polluted basin, Keszthely, compared to the other end of the lake. This gradient is also reflected in the spatial change of the trophic state.

available for algal uptake (for details see Jolánkai and Somlyódy 1981).

(ii) What is the time pattern of the phosphorus release? This is an important issue from the point of view of development, calibration and validation of a dynamic lake eutrophication model (for the present problem see Herodek et al. 1980, van Straten 1980 and Leonov 1980 for the three models under construction) since the exclusion of possible essential temporal changes in the internal loading can lead to false model concepts.

(iii) What is the future behaviour of the sediment after certain external loading reductions? The answer to this question will determine the new equilibrium of the lake and the renewal time needed to reach this state--the most important issue from the point of view of policy making.

Although ecological modeling has advanced considerably during the past decade, the problem of sediment creates a gap in knowledge in this area. This is even more true for shallow lakes (Jorgensen and Harleman, 1978). It is apparent from Leonov's (1981) literature review that there are many approaches (from the simple to the more detailed approach) that describe the phosphorus release, however the possibilities of transferring the results from one system to another are not included in these approaches. The models for Lake Balaton (Herodek et al. 1980, van Straten 1980, Leonov 1980) are also imperfect: the influence of sediment is neglected or incorporated without justification. Thus there is a definite need to improve the part of the model dealing with sediments. The *objective* of this paper is to give background information in this respect.

Sediment biochemistry is a particularly difficult field. There are problems related to sampling, chemical analysis, and laboratory or in situ experiments. The development of detailed models is generally not feasible since the conceptual framework can not be completed with the appropriate information. For this reason the objective here is first to select the major processes and to give order-of-magnitude estimates to them. Only some suggestions will be presented for modeling. As a basis for the current effort, experiments made by Gelencsér and Szilágyi and other scientists have been used.

The paper is organized as follows. First there is a brief general discussion on the major processes (Section 2). The dissolved and solid phases (for the water body and bottom layer respectively) are then characterized for Lake Balaton. In Section 4 the major processes (diffusion, convection, sorption, co-precipitation and resuspension) are selected for Lake Balaton. The order of magnitude (see item (i) above) the spatial and temporal changes (ii) and the possible future behaviour of the sediment (iii) are considered. In the subsequent section, modeling possibilities are analyzed and this is followed by a brief description of the future experimental work required. Conclusions are drawn in Section 6.

2. BASIC PROCESSES

Lakes in general can be considered as heterogeneous, dispersed systems consisting of two phases: in the dissolved phase, the water contains various dissolved materials and ions, while in the solid phase it contains particulate matter. In the water body, the solid phase is dispersed in the continuous dissolved phase; in the sediments the solid phase is more or less compact, the interstices are filled with pore water. The particulate material is generally nonhomogeneous. The interface between sediment and water is not necessarily sharp, it may be quite gradual. However, the ratio of the solid phase to the water phase on either side is drastically different and a sharp gradient exists inside the layer. The change in the ratio is one of the major factors causing differences in the corresponding processes in the water and sediment, and influencing the exchange between them. Another essential difference is that the water body is characterized by assimilative processes (primary production) whereas in the sediment, dissimilative processes (mineralization) predominate (although the latter is also important in the water body).

A further distinction of (i) lake water, and (ii) suspended solids in the water body; (iii) pore water and (iv) sediment in the bottom layer allows a clear indication of major processes influencing the phosphorus release (Figure 1). It is apparent from Figure 1 that sorption can take place in both phases, but a possible adsorption or desorption in lake water is controlled by the amount of sorbent generated by resuspension, and precipitation.

Figure 2 gives more details of the phosphorus cycle. As shown, the processes affecting the phosphorus transformation can be classified as:

(i) biological (primary production, mineralization),

(ii) physical (sedimentation, deposition, resuspension, convection),

(iii) physico-chemical (diffusion, sorption), and

(iv) chemical (co-precipitation, dissolution, reduction, oxidation).

As seen in Figure 2, algae utilizes primarily dissolved inorganic nutrients. These consist mainly of orthophosphate phosphorus, P04-P (or dissolved reactive phosphorus, DRP, see below) as sources of phosphorus (the corresponding sources are carbon dioxide and/or hydrocarbonate, and NO_3^- and NH_4^+ for C and N respectively). After the death of algae, dead organic material (detritus) is formed directly, or indirectly through consumers. The essential portion of detritus is then partly decomposed in water, partly in the sediment layer after deposition. The resulting inorganic P04-P is bound to the inorganic fraction of particulate matter (biogenic lime, co-precipitation, adsorption, etc.) or re-enters the biologic cycle (Figure 2). The oxygen consumption associated with bacterial decomposition often cannot be compensated through oxygen input except in the upper layer of sediment. Therefore, in the deeper zones, anaerobic conditions (i.e. a decrease in the redox potential, Eh) will exist. Generally, the dissolved phosphate concentration in the pore water is much higher than in the water above. This is due to:

(i) The predominance of assimilation in the water and dissimilation in the sediments,

(ii) The lower pH in the sediments as a consequence of this difference, causing CO_2 production and concomitant pH reduction in the sediment. This enhances the solubility of Ca-bound phosphates. In the water body, biogenic lime, precipitation and co-precipitation of phosphates may lower the DRP concentration.

(iii) The lower Eh in the anaerobic part of the sediment will cause reduction of iron and manganese, which further enhances the solubility of the associated phosphates.

The concentration difference in DRP induces diffusive transport from the sediment layer to the water body. A release of dissolved material is also possible due to convection associated with seepage, wind induced interaction and bioturbation.

In addition to the processes previously listed (processes that are typical for most of the lakes) resuspension should be mentioned specifically for shallow lakes. Due to the limited depth, an essential portion of the wind energy input may reach the bottom (in the energy transformation, turbulence and waves play the most important role) and cause an upward flux of sediment and pore water. This flux roughly follows the wind pattern and is characterized by fast dynamics. Wind action causes a release of sediment and total phosphorus bound mainly to particles, but only a limited direct release of dissolved inorganic phosphorus. A more important effect is expected through desorption; the resuspended particles will release phosphorous under changed conditions (pH, Eh, etc.) in the lake water. Biogenic lime co-precipitation leading to the removal of phosphate from the water is a similarly effective, though reverse, process.

3. CHARACTERIZATION OF THE DISSOLVED AND SOLID PHASES FOR LAKE BALATON

3.1 THE SYSTEM OF LAKE WATER AND SUSPENDED SOLIDS

3.1.1 Chemical composition of the lake water

The chemical composition of the lake's water is determined primarily by the major minerals and rocks occurring in the watershed. The high calcium and magnesium bicarbonate content reflects the dolomite composition of the catchment. The longitudinal distribution of Ca and Mg along the lake is peculiar, calcium is highest at the major inflow of the lake in the Keszthely Bay while magnesium shows an opposite tendency (Tóth 1976, van Straten et al. 1979). Approximately 70% of Ca^{2+} that reaches the lake through tributaries co-precipitates. Its extent is closely related to biological processes in the lake (van Straten et al. 1979). The alkaline character of the water is also illustrated by the pH value. This ranges between 8.3 and 8.7 depending on the interrelationship of the primary production and decomposition. If the photosynthetic activity is high, as exceptional a value as pH 9.3 can also be observed. The major anion is HCO_3 (150-200 g/m^3). The conductivity ranges between 480-600 μ S/cm. The large buffer capacity, due to the saturation for $CaCO_3$ in the water explains the small variation in pH.

The dissolved reactive phosphorus concentration (DRP) is determined by the technique of Murphy and Riley (1962) and is generally low, 0.001-0.02 g/m^3. There is no significant change in time or in space. Recent investigations suggest (Istvánovits and Herodek 1981) that the orthophosphate ion differs essentially from DRP and its concentration does not exceed 0.001 g/m^3. As DRP is believed to be completely available for algal uptake, subsequently PO4-P and DRP will be used later in the same sense.

Early model developments for Lake Balaton did not reflect the permanently low DRP concentration. If algal uptake is the only mechanism to eliminate DRP from the water, its temporal change will be ultimately defined by the variation of the primary production resulting in a typical time pattern. As this is not observed in reality, one should assume the presence of some other removing or controlling mechanism such as sorption (see also Oláh et al. 1977, and, in relation to model development, van Straten, 1980).

3.1.2 Chemical composition of suspended solids

The suspended solids concentration ranges in the lake from a background value of 2-5 g/m^3 to 200-300 g/m^3 due to wind action. In the near shore region it may reach 4-500 g/ m^3 (Hamvas 1966, Tóth et al. 1975, Oláh et al. 1977, Györke 1978, Entz 1980). Although suspended solids (SS) are one of the most frequently monitored parameters, information available on them is scanty. According to Tóth et al. (1975) and Oláh et al. (1977) the major fraction of SS is formed by $CaCO_3$ and total phosphorus, TP, is primarily associated with this fraction. The time pattern of TP also reflects well the influence of the wind induced resuspension, however, in comparison with SS, the peaks are perhaps less pronounced.

In 1980 and 1981 investigations were made in order to gain more insight into the composition of the suspended solids, to study the relationship between the concentration (affected mainly by deposition and resuspension) and the composition of SS, and finally to compare the composition of SS to that of the sediment. Emphasis is laid on the analysis of the acid soluble (inorganic) fraction of the suspended solids since this can also be partially mobilized under natural environmental conditions.

Observations were made at two locations, Keszthely and Szemes (Figure 3) representing significantly different trophic states (see van Straten et al. 1979). Samples were taken between 29.08-07.09.1980, seven times in nine days, from three depths at each station. The survey was continued in 1981 with higher spatial resolution but infrequently (twice in 16 points representing the entire lake). From the samples, the concentrations of SS, total particulate phosphorus (TPP), acid soluble phosphorus (ASP), Ca, Mg, Fe and Mn content of acid soluble fractions were determined.

Based on the analysis of data, the following conclusions may be drawn:

(i) The TPP concentration (and also the acid soluble manganese) at Keszthely is essentially higher than at Szemes (from a statistical point of view on a significance level of 1%, see Table 2);

(ii) The ratio of TPP and ASP as an average, ranges between 3.1 and 3.8 for both locations. This means that approximately 30% of the P content of SS is acid soluble;

(iii) In the acid soluble fraction of the suspended solids, the ratio (in terms of weight) of P:Ca and P:Mg are significantly higher for the Keszthely sample compared to the other locations (Table 1). For P:Mn the pattern is the reverse, while for P:Fe no remarkable difference exists. As seen in Table 1, the amount of inorganic phosphorus related to unit Ca weight in the suspended solids of the Keszthely Bay is 2.5 times higher than at Szemes. Observations made in 1981 covering the whole lake showed a 6-7 times decrease in the P:Ca and P:Mg ratios from Keszthely to the other end of the lake (see Figure 4 for the ratio P:Ca). This pattern reflects well the similar longitudinal distribution of the volume-related external phosphorus load (for details see Jolánkai and Somlyódy 1981);

(iv) Assuming that in the suspended solids Ca and Mg in the form of carbonate, Fe as hydroxide while Mn as oxyhydroxide are present 67 ± 15% of SS at Keszthely is acid soluble. The same parameter is 80 ± 9% for Szemes. This ratio was found to be practically independent of resuspension (the SS range was 8-300g$/m^3$) which suggests that the percentages of clay, sand, carbonate, and organic matter in the suspended solids are approximately constant (in other words the sediment stirred up is homogeneous enough). Consequently the sorption properties of the sediment determined by the composition are not affected by storms and can be considered constant on a long term basis.

A summary for the composition of the suspended solids at the two locations is given in Table 2. The comparison between the composition of suspended solids and sediment is discussed later.

3.2 THE SYSTEM OF PORE WATER AND SEDIMENT

3.2.1 Chemical composition of the sediment

The sediment of Lake Balaton is typically a fine sediment: 30-60% of it is formed by magnesian calcite which is mainly of biogenic origin (Müller 1969, Máté 1980). The concentrations of TP and TN range between 0.2-0.6 mg P/g and 2-4 mgN/g respectively (related to dry weight). In TP there is no characteristic longitudinal variation (Tóth 1974, van Straten et al. 1981, Máté 1980) which would reflect the external loading conditions as this component does not express the influence of artificial eutrophication. This is the reason why no significant change exists in the vertical direction in the sediment. There is, however, a pronounced change in the transversal direction (Tóth 1974, van Straten et al. 1979). This can be explained by the increase in the sand content from North to South. Based on extraction analyses, approximately 70-75% of the phosphorus is associated with Ca and Fe (Tóth et al. 1975, Oláh et al. 1977, Dobolyi 1979).

In August 1979 a one year survey was initiated by P. Literáthy which involved four sampling locations (Figure 3, I...IV) in the near-shore region (average water depth approx. 1 m). Sediment core samples were taken once on the first occasion, while pore water samples were taken approximately every month (see Section 3.2.2). Various sediment layers (0-1, 1-5, 5-10 and 10-15 cm) and size fractions (0-63, 63-130, 130-220 and 220-1000 μ m) were distinguished.

Most of the sediment was found in two fractions with the smallest diameter (60-80% in fraction 2 for locations I and II, 40-50% in fraction 1 for the two other locations). The range of organic carbon content is 5-22mg/g dry sediment. As expected, the highest value was found in the upper layer and it decreased by 70-80% in the deepest layer. The C:N ratio exhibits the opposite tendency. Fractions 1 and 4 are the richest in organic material reflecting the planktonic and macrophytic origin. The organic material content is very high in stations I and II being in harmony with the pore water parameters determined (Section 3.2.2). Concerning the spatial variation of TP the same conclusions can be drawn from the results of previous investigators.

As previously mentioned, dissolution of $CaCO_3$ in the sediment takes place due to CO_2 production in the course of bacterial decomposition. This process was modeled on a laboratory scale. It was found that one third of the total phosphorus could be mobilized by CO_2 treatment for the Keszthely sediment. More details are given in Table 3.

The major components of Lake Balaton sediment are listed using Müller's data (see Dobolyi and Bidló 1980) in Table 4. Based on X-ray diffraction and electronmicroscopic analysis, Dobolyi and Bidló (1980) showed the presence of various silicates, magnetite, and rutile as well as brookite. After considerable enrichment they succeeded in proving the existence of hydroxilapatite. Other minerals with phosphorus control were not detectable. Although under the pH conditions of Lake Balaton, the formation of hydroxilapatite is expected, crystallization is impeded by the high magnesium and carbonate ion concentration. The total iron content of sediment ranges between 0.5-3% (in weight).

Gelencsér et al. (1980) found through Moesbauer spectroscope analysis that a portion of the sediment Fe(II) content is associated with chlorite (a clay mineral) and no oxidation takes place after stirring. In the samples taken at Keszthely they succeeded in detecting another Fe(II) composition which can easily be oxidized and probably has a very fine structure. The dominant FE(III) component is the amorphous ferric hydroxide (geothite was not observed). This formation has a large sorption capacity and thus may play an essential role in the phosphorus household of the lake.

3.2.2 CHEMICAL COMPOSITION OF THE PORE WATER

In harmony with Section 2 the pH value of the pore water is significantly lower (7.5 - 8.5) and the electrical conductivity higher (560-800 μ S/cm) than in the lake water (Dobolyi 1980). The DRP concentration ranges in the domain of 50-150 mg/m^2 (Tóth et al. 1975, Dobolyi 1980). Based on Dobolyi's measurements at 11 locations along the longitudinal axis of the lake, there is no essential change in this component and consequently the trophic state of the lake water is not reflected. Generally larger DRP concentrations were found in deeper layers (six layers of 5cm thickness for each were distinguished). According to Oláh et al. (1980) the DRP concentration is between 200-500 mg/m^3 in the 2.5 cm thick upper layer at Keszthely and there is a regular decrease towards the deeper layers (six observations between 26.03-11.11.1980). The difference between the DRP concentrations in the pore and lake water respectively, is one or two orders of magnitude, thus diffusion of importance can be hypothesized (see below).

In the frame of the present investigations, pore water was siphoned out of four locations (see Section 3.2.1. and Figure 3) from two depths (roughly 5 and 15cm). The description of the sampling can be found in Literáthy et al (1981). Components HCO_3^-, NH4-N, P04-P, Fe(II), as well as pH, conductivity, hardness and alkalinity were determined. The summary for the first three parameters is given in Table 5. Exceptionally high values were found for this lake at Keszthely (2 and 16.2 g/m^3 for P04-P and NH4-N, respectively), a clear indication of a sewage discharge nearby. The HCO_3 concentration is the highest at location I. The average value gained for NH4-N is in agreement with the results obtained by Oláh (1980) for the middle zone of the Keszthely bay. In contrast with Dobolyi (1980) a considerable amount of Fe(II) was observed on several occasions. The presence of dissolved iron indicates anaerobic conditions.

The conductivity and dissolved salt content is higher in the pore water than in the lake water due to the excess amount of Ca, Mg and HCO_3. The strong linear correlation found between conductivity, alkalinity and hardness indicates that the dissolution of magnesian calcite is a significant process. The correlation between ammonia and excess hydrocarbonate (above the lake water level) suggests that both components are of organic origin. No significant correlation was found between DRP and any of the other parameters measured. It is suspected that this feature is caused (amongst other things) by nonuniformities in the sediment layer, by the effect of resuspension and by the fact that DRP has a twofold origin: organic and inorganic.

In addition to the experiments outlined, further studies were performed in 1980-81. Sediment was collected three times by an Eckman sampler from 16 points (see Figure 3). At each time and location three samples were taken. Then the upper layer (approximately 5 cm) was homogenized and the pore water squeezed out by N_2 gas. Subsequently various components were determined (DRP, NH4-N, pH, total dissolved iron, etc.) together with the lake water taken simultaneously. It was obvious from the beginning that the Eckman samples are not the most appropriate for an accurate analysis, the aim was simply to get a quick overall pattern for the entire lake. The results are in agreement with previous investigations (Dobolyi 1980, Olåh et al. 1980). Here only the data gained for DRP is presented for later use (See Table 6).

Although the data shows large scattering (partly due to the sampling technique) the resulting high pore water concentration at Keszthely (location 2) is remarkable. Local effects can also be well observed (e.g. location 3 near the mouth of the River Zala and point 13 at the Tihany Strait where the greatest depth is 13m). It is felt that concentrations in Table 6 are underestimations because of the sampling procedure adopted.

In order to get an impression of the short term changes in the pore water quality, daily observations were performed between 20.05-03.06.1981 at Szemes and 04.08-14.08.1981 at Keszthely respectively (see Figure 3 locations 2 and 10). Samples were taken from the upper 5cm layer from 5 points within an area of roughly 50m characteristic size using the modified technique of Literåthy et al. (1980). A lake water sample was also taken (30 cm upper layer). Components such as pH, conductivity, DRP, NH4-N, NO3-N, NO2-N, and total dissolved P, N and Fe were determined. In addition sample chlorophyll, pheophytine and suspended solids, temperature as well as Secchi transparency were measured in the lake water samples. Wind velocity was also recorded. The results are summarized in Appendix I.

At the end of this section we can conclude that the composition of the lake water and pore water respectively, differ essentially from each other, while the same cannot be said for the suspended solids and bottom sediment as the major inorganic components considered. The upper centimeters of the sediment are oxic most of the time, while lower layers are steadily anoxic. No information is available for the organic composition and organic P content of the sediment. Very little is known of the bacterial decomposition in the sediment. However, the available data allows discussion on the quantities of the major mechanisms acting between dissolved and solid phases, such as diffusion, convection, sorption, coprecipitation and dissolution. This is the subject of the subsequent section.

4. BASIC PROCESSES IN LAKE BALATON

4.1 INTERACTION BETWEEN LAKE WATER AND PORE WATER

4.1.1 Diffusion

The difference in concentration between the two water systems induces gradient-type transport tending to equalize this difference. Under pure conditions the flux is proportional to the gradient defined at the interface and the diffusion coefficient, depending only on the temperature of a given material and the classical mathematical description of the process can be used. Under conditions typical for the current problem, several difficulties arise when such a description is adapted. The reasons are as follows:

(i) There is not only a gradient in the constituents (DRP) but also in the diffusion coefficient. The gradient in the concentration is probably more or less inversely proportionate to the gradient in D. Hence the problem is one of measuring a flux and relating this to the appropriate gradient (involving two concentrations and a distance).

(ii) The actual surface area is unknown due to porosity.

(iii) The concentration profile is affected by more factors than diffusion alone, but each factor does not influence the diffusion coefficient. Bioturbation affects D, but sorption and desorption do not. The differential equation governing the problem would have a term for diffusion, convection and reaction. Since, at the interface, reaction may be very important (mineralization, (de-)sorption) the measured (experimental) D may be an artefact (for instance too high, by incorporating implicitly, release in the boundary layer).

The literature (see Leonov 1981) offers a range of $0.6.10^{-6}$ - 4.10^{-4} cm^2/s for the diffusion coefficient. For Lake Balaton two attempts were made to study diffusion. Herodek (personal communication) injected P^{32} to intact sediment core and detected isotope activity as a function of time in the overlying filtered lake water. Dobolyi (personal communication) measured the DRP change in the water under similar conditions (the method of Murphy and Riley, 1962, was employed). No reliably detectable changes were found in the overlying water (in the second case, for example, the technique used could be the reason). The failure of the experiments is probably due to the extremely high reactivity of P; the effect of diffusion is "lost" under all the other transformations. An inert tracer would be better. Thus no conclusion can be drawn from these experiments on the release and the diffusion coefficient.

In the absence of such measurements, the magnitude of the diffusive flux is estimated from existing observations by assuming $0.8.10^{-5}C$ m^2/s average diffusion coefficient. From Oláh et al. (1980) data (six observations) values between 100 and 350 μ gP/m^2d were obtained. This internal load would increase the lake DRP concentration by 1-2 μg/ld. From the daily observations at Szemes (May 1981) and Keszthely (August 1981, see Section 3.2.2 and Appendix I) ranges 20-55 μg/ m^2d and 400-500 μ g/m^2d respectively, were obtained (the gradient was calculated by assuming 5cm thicknesses (perhaps overestimated) for each of the two layers mentioned previously). The flux for Keszthely agrees reasonably with that derived from Oláh's observations and an order of magnitude higher than at Szemes. The explanation for this behaviour is relatively simple. At

Szemes the chlorophyll was surprisingly low (2-7 μ g/l, Appendix I), which is then associated with a presumably low decomposition in the bottom layer causing small DRP concentration and gradient. At Keszthely the temperature was much higher and the Chl concentration (50-100 μg/l) indicated an algal bloom leading indirectly (see Figure 2) to a high DRP concentration in the pore water. It should be noted that from the 16 points survey, a similar range (100-300 μ g P/m^2d) was obtained.

The fluxes derived are relatively small compared to data in the literature (Kamp-Nielsen 1974, Leonov 1981, and others). The reason is the frequent resuspension (typical for wind-influenced shallow lakes) which dilutes the interstitial water on one side and limits the bacterial decomposition in the sediment layer on the other side (consequently there is relatively high decomposition in the lake water). Another reason for the low interstitial concentration and flux could be the low loading compared to other lakes, a high adsorption capacity and a relatively recent history of accumulation. The decomposition in the sediment is quite fast and the phosphate accumulation in the pore water small, thus the gradient formed is also small. The oxygen rich character of the lake's sediment also restricts the development of a high diffusion. Laboratory observations made by Entz (1980) suggest that the release can be 5 to 10 times higher if anaerobic conditions take place. To illustrate the magnitude of the internal load in question it should be noted that the highest estimated value at Keszthely 250-300 kg/d corresponds to approximately half of the external available load (see Section 1).

4.1.2 CONVECTION ASSOCIATED WITH RESUSPENSION

Resuspension of sediment is a typical phenomenon for shallow lakes. During such an event--controlled primarily by wind--not only sediment is released to the water body but also pore water from the corresponding layer. This is a convective type of transport. Somlyódy (1980) showed that the flux of stirring up is approximately linearly-related to the wind speed (see Section 4.3). Assuming 75% water content in the sediment layer and 150-200 μg/l DRP average concentration in the pore water 10.W [kg/d] can be gained as a first estimate for the release for the entire lake (W is in m/s). Taking 5m/s average speed this leads to 50kg/d, 10% of the lake's external available phosphorus load.

It is suspected however that, due to wind action, a thicker layer is loosened in the sediment than is stirred up (around 1mm) and that pore water can be supplied from this region to the lake. Thus the release can be several times larger than estimated if the upper part of the sediment is evenly rich in DRP. The depth of the mixed sediment layer can be estimated on the basis of the vertical distribution of the chlorophyll content in the sediment. As Szilágyi (1981) showed, in the upper 2cm, the Chl-a content is higher by one order of magnitude than in lower regions, this assumption should be accepted. Observations made by Oláh et al. (1980) also support this tendency. It should be noted that the associated concentration increase in the lake water is near to the detectable level even when all the uptake mechanisms are neglected: a clear indication why no DRP increase is observed during storms (see for example van Straten et al. 1979). It is also worth noting that, based on the limited information available, seepage seems to be negligible in this case (Major, personal communication).

4.2 INTERACTION BETWEEN DISSOLVED AND SOLID PHASES

In this section sorption, chemosorption and coprecipitation-dissolution are discussed. Chemosorption, being mainly irreversible and with a time scale which is different from (physical) sorption, is treated together with coprecipitation.

4.2.1 SORPTION

The adsorption properties of the phosphate ion are discussed by various authors especially for solid phases which formed by iron, aluminium and calcium (see for example Golterman 1975, 1980, Lijklema 1980). For Lake Balaton among components of sediment and suspended solids (see Section 3) first of all, magnesian calcite and amorphous ferric hydroxide should be mentioned due to their high adsorptive capacity.

According to Jacobsen (1978), the adsorption of phosphate on calcite in the relevant pH range of 7-9 is essentially independent of pH, but increases with a higher magnesium content of the calcite. The sediment of Lake Balaton presumably has favourable sorption properties since small fractions (< 63 μ m) with a high specific surface are dominant (Müller 1969, Mátė 1980). According to Mátė's systematic observations in space the mean particle site ranges between 10-20 μ m in Keszthely and Szigliget (nearly 100 samples were taken) except the southern shoreline where the sand fraction is more dominant. Györke (1978) found similar characteristics, both for the sediment and suspended solids.

The sorption capacity of amorf ferric hydroxides formed in the course of weathering of iron minerals in the water environment is influenced by pH and redox potential (Eh), but ageing and re-crystallization also play an essential role (Lijklema 1981). The increase in pH decreases the sorption capacity and this change is apparent in the domain 7-9. Under reduced conditions the dissolution of iron and consequently the release of phosphate can be typical.

For Lake Balaton two features are expected to be of major importance:

(i) The sorption equilibrium is formed quickly both in the lake and pore water according to the corresponding DRP concentrations and pH values; and

(ii) In the course of resuspension, desorption of calcite and amorf ferric hydroxide is the dominating process, due to the change in the DRP concentration (and perhaps in pH).

The first sorption experiments (Tóth et al. 1975) showed a high adsorption capacity, 200 μ gP/g dry material (it should be noted that the suspension employed had an unrealistically-high concentration of 50g/l). The desorption studies resulted in the pH range 8.5-5.0 values 50 μ gP/gDM and 1.6 μ gP/gDM for Keszthely and Szemes respectively (DM dry material). A minor dependence on pH was also found. A sorption isotherm cannot be derived from the data.

In the frame of the present investigations both adsorption and desorption studies were performed. As a first step, the influence of the major parameters (initial DRP concentration, suspended solids concentration (SS) and reaction time) was surveyed (see Table 7). A sample was taken from the upper 3cm layer with Eckman equipment from the Szemes basin. The DRP concentration in the pore water was 179 μ g/l while the pH and conductivity 8.2 and 600 μ S/cm respectively. The same parameters for the lake water are as follows: <1 μ g/l, 8.7 and 520 μ S/cm. Based on the results (see Table 7) the following can be stated:

(i) Depending on the initial concentration, both adsorption and desorption take place;

(ii) The amount of sorbed material depends on SS and the reaction time. Adsorption seems to be a slower process compared to desorption which is practically completed within 30 minutes (Figure 5);

(iii) The pore water concentration, 171 μ g/l might not be an equilibrium concentration as the resuspension of this sediment in water with 150 μ gP/l resulted in net adsorption although desorption would have been expected. The other explanation may be that the higher (apparent) equilibrium concentraton of PO4-P in the pore water (171 kg/l, if correctly measured) is due to the higher concentration of ions (including HCO_3 which competes with HPO_4^{2-}) compared to the lake water.

In order to analyze sorption in more detail, sediment was collected from various regions of the lake representing different trophic states and laboratory experiments were performed. The adsorption tests are discussed first.

ADSORPTION

Sediment was taken from Keszthely and Szemes. 5-7 suspension solutions were prepared (three parallels for each) covering a range of 0.3 to 8.5 g/l. Lower values are realistic during storms while the higher concentrations may occur only near the bottom. For preparing the suspensions, River Zala water with its natural DRP content (range 150-310 μ g/l) was used. Beside continuous stirring the decrease in the DRP concentration was monitored (it is worth noting that parameters such as pH and temperature were not measured and the major characteristics of the sediment are unknown). Results for the Keszthely Bay are illustrated in Figure 6 where the percentage of adsorped phosphate (related to the initial concentration) is given as a function of SS and reaction time. As seen in the figure, the process is quite fast at the beginning but, after an hour, the removal becomes less extensive. Within 3 hours equilibrium is reached only for large SS concentrations but still the tendency of the results suggests the existence of a reasonable isotherm (it should be noted that for the 0.3-1.2 g/l SS domain a sample was also taken after about a day, see below). The adsorption capacity represents a similar magnitude (15-150 μ g/gDM, higher values belong to lower SS concentration) and hence to raise the ultimate DRP concentration--as found by Tóth et al. (1975).

Subsequently, adsorption tests were performed in lower and more realistic ranges of DRP and SS. Sediment samples were taken from the middle of the Keszthely, Szigliget, Szemes and Siofok bays (Figure 3). Suspensions were established in the 50-2000 mg/l SS domain with filtered lake water and the initial DRP concentration increased to the level of 32 μ

g/l. Based on the experiences of the previous experiments, a reaction time of 24 h was chosen. The adsorped amount of phosphate per unit weight sediment (Δ C/SS, Δ C = C (0) - C(24)) is illustrated in the function of the equilibrium concentration, C_e (approximated by C(24)) in Figure 7.

All the points together serve a scattered picture. If, however, the individual samples are considered separately, an isotherm-like trend is more apparent. The Szigliget sediment has the highest adsorption capacity. The Siofok sediment the smallest, while the two other types represent the same medium range. It is stressed that the results are subject to great uncertainty due to errors in the DRP and SS determinations. In the figure the uncertainty related to the analytical error in DRP (δ c) is illustrated for the Keszthely sediment. This uncertainty is especially high for low sediment concentrations when the absolute removal is small and the magnitude of Δ C is the same as that of δ C. An upper limit of the adsorption capacity is indicated by a broken line in the figure.

Figure 8 indicates the domains of both adsorption experiments discussed previously. The dots represent results from the first test (Keszthely, SS = 0.3-1.2 g/l, average of three parallels). As can be seen, the two experiments coincide with each other.

It is worth noting, that Hieltjes (1980) found similar adsorptive capacity for Brielse Meer sediment which is also calcium rich, but at much higher DRP concentrations. This suggests that the Lake Balaton sediment has a very high adsorptive capacity.

DESORPTION

For this study the same sediment samples taken at Keszthely, Szigliget, Szemes and Siofok for the adsorption study were used. Suspensions were prepared with filtered lake water without adding phosphate ion to the system. Based on previous experiments (Figure 5) half an hour reaction time was adopted. The results are summarized in Table 8. As can be seen, desorption was found in all cases.

As indicated in Figure 9a the sediment is removed from its original equilibrium (1) and taken to a new environment (2) characterized by a much lower DRP concentration (and also different pH, conductivity, etc.). In the course of the stirring experiment a new equilibrium (3) is reached through phosphorus release from the sediment. The concentration increase caused in the water, Δ C, is monitored and the change in the adsorped amount of phosphate, Δ C/SS, is derived (see Figure 9a). It should be stressed that changes along both axes are determined from a single measurement, i.e. no independent checking exists.

The desorped phosphate per unit sediment weight calculated from the data of Table 8 is plotted against the new equilibrium concentration in Figure 9b. The uncertainty ranges, as calculated previously, for adsorption from the three parallel measurements are also given. From the figure, the following conclusions can be drawn.

(i) As an average 5-10 μ gP/gDM desorption is obtained. With a moderate storm leading to 25 mg/l average increase in suspended solids, a release of 250-500 kgP is expected, the same order of magnitude as the external load.

(ii) Although the presence of desorption is clearly indicated, the large scatter and uncertainty of points associated with analytical difficulties discussed earlier do not allow the establishment of a Δ C/SS = f(Ce) relationship. Some points (indicated by question marks) are suspicious. The expected trend of the data is illustrated by an approximate envelope (broken line). The analytical technique should be improved in order to obtain more reliable results.

(iii) Δ C/SS = f(Ce) is not equivalent to the isotherm given in Figure 9a. In order to get the latter, Δ C/SS should be subtracted from (P_{ad})max (the maximal amount of P fixed by sorption/unit sediment weight) which is unknown (in fact the particles' surface related concentration is not known). The total P concentration of the sediment ranges between 200-600 μ g/gDM, certainly a much higher value than (P_{ad})max.

It should be noted that desorption experiments were also performed twice with sediment collected from 16 locations of the lake (see above). The SS concentration was about 6g/l and the reaction time 24 h. The equilibrium concentration ranged between 30-50 μ g/l and the desorption capacity in 5-10 μ gP/gDM. The results are in agreement with the trend in Figure 9b.

The pH dependence of the process was also studied. No influence was found which can be explained by analytical difficulties and presumably by the ratio of Ca/Fe (between 40 and 80). Further efforts are required in this respect.

4.2.2 CO-PRECIPITATION AND DISSOLUTION

In the lake 3.4.10^4t Ca^{2+} is stored annually. The average primary production is approximately 830gC/m^2 yr for Keszthely and 96gC/m^2 yr at Siófok while the associated formation of $CaCO_3$ ranges between 800-7000 g/ m^2 yr (van Straten et al. 1979). Assuming that during biogenic lime production the P concentration is constant in the suspended solids and the Ca:P ratio is 200 and 1000 for Keszthely and Siófok respectively (see Section 3.2), the amount of co-precipitation phosphorus is around 14gP / m^2yrand0.3gP / m^2yr for the two basins. The difference between the two estimates shows the extremely important role of algal activity in removing the major portion of phosphorus reaching the lake through the River Zala at Keszthely by an inorganic chemical process. The above values are overestimated since no decomposition was assumed in the water column. Using the overall ratio 1:2 for mineralization (sediment:water, see previous section) 4.7 and 0.1 gP / m^2yr are arrived at, to which 12.9 and 0.27 mgP / m^2yr for for the entire lake, 1800 kg/d removal is obtained, illustrating the importance of the co-precipitation in the phosphorus cycle of the lake (note that more than 90% of this amount is then accumulated in the sediment). The accuracy of the estimate given is influenced by various factors such as the real Ca:P ratio in the freshly co-precipitated biogenic lime (the ratio found in the suspended solids reflects rather a long term average), the CO_2 production associated with bacterial activity, etc.

It is stressed that co-precipitation strongly influences all the sediment related processes. Dissolution of $CaCO_3$ in the sediment counteract the incorporation of P in the solid phase and resuspension can lead to desorption in the water body. In a simplified way, it can be stated that co-precipitation is the really effective "adsorption" counterpart of desorption. The balance between the two processes takes place, not directly, but through the biological, chemical and physical processes; it is illustrated in Figure 2.

4.3 WIND-INDUCED RESUSPENSION

With a certain desorption capacity (see Section 4.2.1) the release of phosphorus depends solely on the amount of resuspended sediment. Thus resuspension plays an extremely important role. It can be described in three stages:

(i) The establishment of hydrophysical parameters characterizing the shear stress or energy conditions near the bottom;

(ii) The computation of these parameters as a function of wind characteristics, geometry, fetch, etc. Both empirical approaches and more detailed hydrodynamic models can be used (Lam and Jaquet 1976, Sheng and Lick 1979, Fukuda and Lick 1980). If the contribution of wave motion to the shear or energy at the bottom is essential some wave hind-casting method should be employed;

(iii) The elaboration of a relationship between hydrophysical parameters and sediment release which strongly depends on the composition of the sediment. Mainly laboratory experiments are used here (see for example Fukuda and Lick 1980).

For Lake Balaton, the early observations of Hamvas (1966) and Györke (1978) showed the close relationship between wind and SS. Entz (1980) showed that the response of the system due to the shallow water is very fast. A more comprehensive study was done by Somlyódy (1980, 1981). The original aim was to identify the flux of resuspension (together with sedimentation) from regular in situ observations through an appropriate (simple) model development. Water samples were taken at the middle of the Szemes basin (water depth H = 4.3 m) from five depths between 15.05-31.10.1979 once each day and the SS concentration determined (several other components were also measured). Wind speed and direction were recorded continuously. The model development started from a vertically one-dimensional, unsteady transport equation for SS which was then integrated along the depth. The simplified ordinary differential equation (ODE) involved the two opposite fluxes for the deposition and resuspension, respectively. For the fluxes, hypotheses were made; e.g. for describing resuspension, a simple energy transformation principle between potential energy and turbulent kinetic energy was adapted (a similar method is often used when calculating thermal stratification for deep lakes). Thus an ODE with three parameters was arrived at for the depth average SS concentration, C

$$\frac{dC}{dt} = \frac{1}{H}\left(\varphi_d - \varphi_c\right) = -K_1 C + K_2 W^n \qquad 1$$

where φ_d and φ_s are the two fluxes and W is the absolute value of the wind velocity vector. The unknown parameters were estimated based on the time series C(t) and W(t) gained from observations. Both a deterministic least square fitting technique and the Extended Kalman Filter (Beck and

Somlyódy, 1982) were employed. The latter method was also used for model structure identification and validation.

For the two fluxes equations

$$\varphi_d = 5.6C \;\; kg/m^2d \tag{2}$$

and

$$\varphi_e = 0.034W \;\; kg/m^2d$$

were obtained (n = 1). The coefficients represent physically realistic values (e.g. 5.6 m/d for the sedimentation velocity, $C_e = 0.6.10^{-2}W[kg/m^3]$ for the steady state concentration).

As is apparent from Equation 1, the change in C depends on the relative magnitude of φ_d and φ_e. From the point of view of desorption however, it is not this change, but the amount of freshly released sediment φ_e, that is of importance. Consequently a higher internal load is expected of them from the example given in Section 4.2.1 if the sorption properties are constant within the layer subject to subsequent resuspensions (this is a reasonable assumption as the stirred layer is very thin). Assuming again 5-10 μg P/gDM desorption capacity as a first estimate (100-200)W kg/d load is derived for the whole lake. Thus values of about 1000 kg/d can easily occur.

It should be noted that the constant in the equation of φ_c is inversely proportionate to depth. Thus, for an average basin with depths deviating from 4.3m, the relation

$$\varphi_s = 0.034\left(\frac{4.3}{H}\right)W \tag{3}$$

should be used.

Several methods exist for improving the existing resuspension models. For instance, the utilization of the daily observations (a six week period) of the first two authors for Keszthely and Szemes and other measurements available (Hamvas 1966, Györke 1978, Entz 1980, Máté 1980), or the further development of the hydrophysical part of the model can be mentioned (see Somlyódy and Virtanen 1982). However, it is felt that the present model version is appropriate enough and no modification is needed unless the description of the sorption is essentially refined.

4.4 ORDER OF MAGNITUDE ESTIMATES: SPATIAL CHANGES AND DYNAMICS

In the previous sections three release mechanisms: diffusion, convection and desorption, and a removal process: co-precipitation were discussed (adsorption is felt to be of minor importance, see Section 4.2.1). Among the release processes, desorption is the most effective, it can lead to an internal load of 1000kg/d depending on wind and the chemical environment. From the data, no conclusion can be drawn for the spatial changes. The contribution of diffusion seems to be smaller, 250-300 kg/d at Keszthely with a decreasing tendency towards the eastern end of the lake. Convection associated with wind action shows the same trend and may have the same magnitude as diffusion. Co-precipitation has a comparable magnitude to the sum of convection, desorption and diffusion or, in a different way, it can roughly balance the three load terms. It is quite difficult to gain a close pattern in the temporal changes. Existing data do not show appreciable fluctuations in the DRP concentrations; thus the counteracting processes of diffusion, (ad-) and (de-) sorption, mineralization and algal uptake should be more or less balanced. Further experiments (dynamics of bacterial decomposition, formation of the gradient, diffusion coefficient, etc.) would be required to get insight into the temporal variations. The same is true for convection since it also depends on the pore water concentration. Wind certainly causes a noisy pattern but on a weekly or monthly basis--changes for such periods are really important from the point of view of the entire eutrophication process--the fluctuations are practically smoothed out. The situation is similar for desorption since it depends in the same way on wind. A major effect is caused however by the shape of the sorption isotherm. With decreasing lake water phosphate concentration, the desorption capacity increases quite as much, especially in the small concentration range--thus essentially influencing the time pattern of the desorption flux. At the same time the uncertainties in the isotherm can result in large errors in estimating the release associated with desorption. The dynamics of the only sink process--co-precipitation--tackled here is determined by primary production of algae and bacterial decomposition.

4.5 FUTURE BEHAVIOUR OF SEDIMENT

Our knowledge is very limited in this central issue irrespective of the type of system considered (lakes, reservoirs or issues) or the nature of the problem (eutrophication, heavy metal pollution, etc.). In most of the cases nothing more than speculation can be put forward.

To start with, it is worth considering briefly the past behaviour of the sediment in the Balaton problem. It can be assumed that during the first period of man-made eutrophication (until the early seventies) the phosphorous which had accumulated in the sediment became inactive through various removal mechanisms (mainly adsorption of iron and biogenic lime co-precipitation corresponding to actual primary production). With the gradual increase of the external load, however, the production and decomposition also further progressed and the sorption capacity of amorphous ferric-hydroxide reached saturation, leading to a considerable increase in the internal load and greater deterioration in the water quality.

Concerning future behaviour (third period) two alternatives are discussed:

(i) Production, decomposition and the internal load will increase further. An acceleration of the eutrophication is foreseen through the appearance of anoxic conditions resulting in the re-mobilization of phosphorous fractions buried in the past. The internal load can essentially exceed the external one and phosphorous may not be the limiting factor of eutrophication anymore. The reduction of the external load at this stage is far less effective than around the second period.

(ii) Qualitatively it can be stated that a reduction in external loading of phosphate will reduce the primary production and the lime precipitation. As a consequence, less organic material, $CaCO_3$ and phosphorous will precipitate, not in proportion however, but with a higher percentage reduction in net phosphate because not more P can precipitate than imported, but the production of organic matter and $CaCO_3$ can be sustained by recycling of nutrients (internal loading). Because other constituents will continue to deposit (clays, sand, humics) the sediment will dilute in P and in organic matter. Anaerobic conditions will be reduced in time and space; this fact will also contribute to a gradual reduction in internal loading. In turn, a further reduction in productivity is obtained. Based on our present knowledge, it is not possible to offer a quantitative picture and to calculate the removal time of the sediment (and consequently that of the lake). Some estimate of future conditions can perhaps be obtained by the analysis of historical data (also requiring the history of the external load) and/or longitudinal variations, as both reflect different P-loading of the sediment.

At the end of this section it is noted that the magnitude of regeneration time, depending on the character of the lake, is 5-10 years. This time increases as the internal load increases (in relation to the external use). From this, an important practical conclusion follows: one year's delay in reducing the external load will take more than two years to achieve the same water quality level in the lake. Thus the effectiveness of the same management action can strongly diminish as time passes.

In summary it should be stressed that the characterization of the dynamic properties need further experimental work simultaneously with model developments which are capable of putting together the various processes discussed here and indicated in Figure 2.

4.6 BEHAVIOUR OF THE SEDIMENT AFTER EXTERNAL LOADING REDUCTION

When discussing this crucial question it is worth following the previous behaviour of the sediment. It is believed that, during the first period of artificial eutrophication (perhaps until the beginning of the seventies) an essential portion of phosphate reaching the lake was removed by adsorption of iron and by biogenic lime co-precipitation corresponding to the algal production at that time. With a growing external load, the primary production, bacterial decomposition, and, as a result of dissolution, the internal load, all increased.

Presumably the high-energy adsorption sites of the amorphous iron (III) hydroxides gradually become saturated, which causes the iron-phosphate equilibrium to shift towards higher equilibrium phosphate concentrations. Such transitions progress gradually from west to east in the lake. In addition, the increased organic loading results in a higher seasonal variation in the aerobic sediment layer and subsequently in higher release rates during the summer periods. After reducing the external load, more or less the opposite changes are anticipated. Since the internal load has the same magnitude as the external one, algal production can remain unchanged for a while. The biogenic lime co-precipitation is however decreasing, thus leading to a "dilution" of the sediment in phosphate. The time scale of this process is of primary importance as this will be reduced through the decrease of the DRP concentration in the pore water diffusion and convection. In addition, a shifting of the nominal point along the adsorption isotherm and a possible modification of the isotherm is determined by this phenomenon (see for example Lijklema 1981).

5 MODEL DEVELOPMENTS AND FUTURE EXPERIMENTS

5.1 MODEL DEVELOPMENTS

The importance of modeling has already been stressed in the Introduction. In the light of Section 4, the definite need for modeling became apparent from another side: the information available is not sufficient to adequately describe the isolated subprocesses discussed here. Therefore, perhaps the only possible way of developing hypotheses at present--based on our existing knowledge--is to incorporate them in the frame of the lake water quality models under development and test the hypotheses using the estimates given here (and other data used previously in the modeling framework). This procedure can hopefully lead to an improvement of the models in question from the side of the internal load which was the objective of the present effort. Subsequently, a brief discussion is given on the modeling of the various subprocesses related to the sediment.

5.1.1 DIFFUSION

The simplest possibilities are as follows:

$$\varphi_{diff}(t) = const;$$

$$\varphi_{diff}(t) = k_1\left[k_2 - DRP(t)\right].$$

where in fact k_2 is the DRP concentration of pore water considered constant in time and both for k_1 and k_2 relatively well defined ranges can be obtained from Section 4.1.1;

$$\varphi_{diff}(t) = k_1\left[DRP_2(t) - DRP_1(t)\right]$$

the same expression as before but with time variable pore water concentration. In order to describe this latter the decomposition of detritus in the sediment should be included (otherwise it is sufficient to assume that complete decomposition takes place in the water body, see for example van Straten 1980).

It is felt that a constant additive load term would not improve our understanding of the system's behaviour. Thus option (ii) or (iii) should be realized. The advantage of (ii) is its simplicity. Expression (iii) may characterize better the dynamics of the process, but the price for it is the appearance of a new state variable, one to three additional parameters and an initial condition in the model. Still the test of hypotheses listed is simple enough.

5.1.2 CONVECTION

Wind is suggested to be incorporated as a daily or weekly average. For $DRP_2(t)$ the same procedure can be employed as before in harmony with the approach chosen for the diffusion.

5.1.3 SORPTION

An obvious choice is to assume a Langmuir isotherm (as a first step assumed to be valid equally for both the lake and pore water),

$$P_{ad} = (P_{ad})_{\max} \frac{Ce}{k_1 + Ce},$$

where both k_1 and $(P_{ad})_{\max}$ should be considered as unknown parameters (still with reasonable ranges gained from observations). At least two basic cases exist depending on the character of the change in the system (Figure 10):

(i) Resuspension

The resuspended sediment which was originally in equilibrium reaches a new equilibrium (figure 10a, point (3)) in the lake water as discussed in Section 4.2.1. The new position on the isotherm can be calculated from the simple condition that concentration increase in the lake water is derived from the phosphate release of the resuspended amount of sediment. In other words, point (3) is defined by a straight line originating from point (2) the gradient of which is inversely proportionate to the amount of sediment stirred up (or to SS). Thus for a small SS, (3') is obtained while for a large value of SS (3'') is obtained (see Figure 10a). The resuspension itself can be calculated according to Section 4.2.1. It should be noted that the presence of suspended solids in equilibrium in the water before resuspension (see point (4)) can be easily taken into account in the course of the computation.

(ii) Sudden external load increase for originally equilibrated lake water-suspended solids system

Adsorption will take place as indicated in Figure 10b which similarly depends on the SS concentration as in the previous case. If resuspension occurs at the same time, the "old" suspended solids will adsorb while the fresh ones desorb. The new equilibrium can also be derived without difficulty in this situation.

The modeling procedure outlined is simple and straightforward. Difficulty is caused by the fact--as previously stressed--that the computations should be performed primarily in the 0-20 μg/l domain where the isotherm is most uncertain and has the highest gradient. Thus special care should be taken in the course of the modeling.

5.1.4 CO-PRECIPITATION

For possible alternatives the reader is referred to Herodek et al. (1980) and van Straten (1980).

5.1.5 MODELING THE FUTURE BEHAVIOUR OF THE SEDIMENT

Based on Section 4.5, the analysis of historical and longitudinal changes may serve as a basis for model development. It would require the following steps:

(i) the establishment of the load history for the last 20-25 years;

(ii) the improvement of the sediment compartment of the existing lake models as suggested in this section; and

(iii) running the model for the 20-25 year period and using historical in-lake water concentrations for calibration.

Such a procedure would help avoid definition of some of the essential parameters which determine the long term behaviour of the sediment (e.g. initial concentrations) since the changes are followed right from the (approximate) beginning of the process. It is admitted however, that considerable effort is necessary in order for the approach outlined above to be realized and its succes cannot be guarantied because of the various existing uncertainties (e.g. the reliability of load history).

5.2 FUTURE EXPERIMENTS

It is not the intention here to give a detailed list for future research since the need ultimately follows from Section 4. It should be noted however that both the spatial and temporal resolution of the sampling should be refined. Special care should be taken of diffusion, but first of all the proper determination of the adsorption isotherm and experiments related to the biogenic lime co-precipitation have to be mentioned. It is felt that changes in analytical techniques are also required, e.g. the development of a more sensitive method is the pre-condition for obtaining isotherms that are sufficiently accurate.

6. CONCLUSIONS

1. Most of the sediment in Lake Balaton can be found in the fraction 0 - 130 μm. Its water content is approximately 70%. The sediment is much finer along the northern shoreline than near the southern shoreline where its character is sandy. The sediment consists mainly of carbonates (up to 65%), sand (30-40%) with some iron hydroxide and a fairly low content of organic matter. From the viewpoint of phosphate binding carbonates and iron hydroxides are the most important. The total phosphorus concentration, TP, ranges between 0.2 and 0.6 mgP/g, roughly half of which can be extracted by acids. The longitudinal change of TP does not reflect the pronounced decrease of the external load from west to east, but there is an apparent transversal variation. Most of the phosphorus can be found in the 0 - 63 μm fraction. While the composition of the suspended solids and the sediment are similar, the pore water differs essentially from the lake water. It has a much lower pH (range 7.5-8.5) and a higher dissolved reactive phosphorus concentration (100-300 μg/l as compared to 2-10 μg/l). In the vicinity of sewage discharges concentrations up to 2000 m/l were measured, but otherwise the spatial variation is not pronounced (a modest decrease can be observed from west to east). Because of the strong wind action and low water depth, anoxic conditions occur only occasionally.

2. The external load of the lake in terms of total phosphorus is about 1000 kg/d (Jolánkai and Somlyódy, 1981), half of which is considered available for algal uptake. The phosphate release from the sediment, called the internal load, now represents the same order of magnitude as the external load.

3. The dominating process in the phosphate release is desorption, representing approximately 1000 kg/d load on average. The contribution of diffusion on a lake wide average is 250 - 300 kg/d, while the third mechanism, convection of pore water due to wind action, causes a smaller effect. The really effective counterpart of desorption is the biogenic lime co-precipitation which reduces the influence of the source terms listed above.

4. The dynamics of desorption are fast, on account of the wind induced resuspension, (but on a weekly or monthly average the changes are not so pronounced for a year) while diffusion shows less variability. The wind is of major importance in phosphate release and the situation can be similar for other shallow lakes.

5. A number of sorption experiments were performed. The methods used are, however, not accurate enough to determine the sorption isotherm properly (especially not in the initial domain where the gradient is the largest). Further efforts which would change the analytical techniques employed would still be required. Experiments are also needed in relation to biogenic lime co-precipitation.

6. On the basis of the research discussed here, quite straightforward suggestions were made on how one could incorporate the sediment processes in ongoing developments of eutrophication models of Lake Balaton.

7. One of the most important issues from the viewpoint of lake eutrophication management is the future behavior of the sediment after load reduction. Only vague statements can be made in this respect. The

utilization of historical data and the presence of spatial changes in the system are suggested here. This could be done in a modeling framework when the changes in the lake and sediment are simulated from the "beginning" of artificial eutrophication. The procedure requires the establishment of the load history and special care should be taken to account for the various kinds of existing uncertainties (in the load estimate, in observations, etc.).

Table 1. Ratios of P/Ca, P/Mg, P/Fe and P/Mn in the acid soluble part of suspended solids at Keszthely and Szemes (29.08.-07.09.1980, ± relative standard deviation in percentage from 20 data)

Sampling location	P/Ca	P/μg	P/Fe	P/Mu
Keszthely	$2,44.10^{-3}$ ± 22%	$2,94.10^{-2}$ ± 21%	$8,70.10^{-2}$ ± 29%	$3,94.10^{-1}$ ± 22%
Szemes	$1,11.10^{-3}$ ± 21%	$1,49.10^{-2}$ ± 30%	$7,04 \pm 10^{-2}$ ± 51%	$5,99.10^{-1}$ ± 20%

Table 2. Chemical composition of the suspended solids (W%)(29.08-07.09.1980)
K - Keszthely, 19 data
Sz - Szemes, 20 data

		TPP	Acid soluble				
			P	Ca	Mg	Fe	Mn
mean	K	0.22	0.059	23.51	1.98	0.66	0.15
	Sz	0.11	0.033	29.54	2.19	0.47	0.056
variancia	K	32.0	25.4	22.1	27.7	28.2	32.4
%	Sz	29.5	21.2	13.2	19.2	48.9	14.3
minimum	K	0.1	0.04	8.51	1.02	0.20	0.05
	Sz	0.03	0.02	23.21	1.51	0.08	0.04
maximum	K	0.34	0.11	31.26	2.94	1.09	0.22
	Sz	0.16	0.05	37.88	3.10	0.89	0.07

K - Keszthely, 19 data
Sz - Szemes, 20 data

Table 3. Carbon dioxide extracted phosphate content of the sediment

Sampling locations*	CO_2 extracted		TP	$(PO4\text{-}P/OC)$**
	$(Ca,Mg)CO_3$ mg/g	PO4-P µg/g	µg/g	mg/g
I				
0-5 cm	39.9	50	380	2.8
10-15 cm	19.9	33	320	3.8
II				
0-4 cm	123.0	120	310	2.2
30-40 cm	155.0	22	350	0.3
Keszthely midpoint				
0-5 cm	320.0	150	450	1.1
III				
0-4 cm	32.0	29	190	2.1
4-10 cm	53.0	44	180	1.9
IV				
0-3 cm	47.0	57	310	2.7
6-10 cm	28.0	5	150	0.4

[*] see Figure 3.

[**] amount of phosphate mobilized from sediment by 1g mineralized organic carbon

Table 4. Substantial components of Lake Balaton sediment after Müller 1969, Dobolyi and Bidló 1980 (rare compounds are in brackets)

Allochthone components	Autochthone components
"Sand components"	Mg-Calcite
Quarts	(aragonite)
Rock debris mainly lime and dolomite	
Feldspars (potassium feldspar, Plagioclase)	Opaline silica
Mica (predominantly muscovite)	
Chlorite	
"Clay components"	
Montmorillonite	
Illite	
Kaolinite	
Organics	Organics

Table 5. Concentrations of nutrients in the lake water (lw) and in the pore water (pw) near the shore (08.1979-06.1980)

Location (Fig. 3)		HCO_3^- [g/m^3]				NH_4-N [mg/m^3]				PO_4-P [mg/m^3]			
		mean	Std. dev.	min.	max	mean	Std. dev.	min	max	mean	Std. Dev.	min	max
I	lw	262.3	30.5	201.3	298.9	224	322	<14	742	6	9	<3	31
	pw	695.4	237.9	305.0	1110.2	8330	6342	714	20720	189	171	37	639
II	lw	268.4	42.7	201.3	341.6	126	98	<14	266	25	39	<3	130
	pw	506.3	146.4	298.9	738.1	7308	5068	1498	16240	673	527	118	1919
III	lw	274.5	48.8	201.3	341.6	168	238	<14	602	22	34	<3	102
	pw	396.5	97.6	286.7	579.5	3822	3472	84	10640	195	164	<3	549
IV	lw	268.4	12.2	244.0	286.7	98	84	14	238	3	3	<3	16
	pw	439.2	103.7	347.7	616.1	4060	3640	84	11060	37	31	<3	96

Table 6. Spatial distribution of DRP concentration in lake water (lw) and pore water (pw) (note sediment was collected by Eckman sampler)

Date / Location (see Fig. 3)	11.09.1980		6.04.1981		22.06.1981	
	pw	lw	pw	lw	pw	lw
1	65	-	253	16	229	7
2	274	0.013	300	4	222	8
3	224	-	280	5	114	22
4	47	-	163	8	54	7
5	37	0.012	115	7	63	7
6	60	-	84	5	195	9
7	4	0.009	153	5	51	5
8	58	0.003	123	7	50	7
9	70	-	217	5	53	7
10	36	0.008	101	18	34	5
11	50	-	88	9	72	7
12	78	0.001	231	2	75	7
13	28	0.001	555	7	58	5
14	27	-	177	1	182	5
15	4	0.001	271	6	37	5
16	15	-	123	1	40	2
17	18	0.001	-	-	-	-

Table 7. Influence of major parameters on sorption (sample was taken at 21.05.1981)

No. of experiments	Reaction time [hour]	Suspended solids concentration [g/l]	Initial PO-P concentration [μg/l]	Final PO-P concentration [μg/l]	Sorbed amount of P** [μgP/gDM]
1	3	5.9	132	53*	+132**
2	3	5.9	< 1	43	- 7.2
3	3	0.59	132	93	+65.0
4	3	0.59	< 1	25	-41.7
5	1	5.9	132	82	+ 8.3
6	1	5.9	< 1	40	- 6.7
7	1	0.59	132	130	+ 3.3
8	1	0.59	< 1	14	-23.3

*Average of 3 parallel observations

**negative value means desorption

Table 8. Results of desorption experiments (28.10.1981).
Reaction time 30 min.

Keszthely		Szemes	
SS [mg/l]	C_e [µg/l]	SS [mg/l]	C_e [µg/l]
71.9	3.6 ± 1.0	122.1	3.3 ± 0.6
83.1	3.0 ± 0.7	391.2	4.9 ± 0.3
230.8	4.1 ± 0.3	751.6	7.1 ± 0.6
230.7	4.7 ± 0.0	1035.9	7.6 ± 1.6
553.9	6.6 ± 0.3	901.5	9.1 ± 0.6
1305.5	10.2 ± 0.3	2060.5	16.1 ± 7.5
lw	2.4 ± 0.3	lw	3.4 ± 0.9
Szigliget		Siófok	
90.3	2.4 ± 0.2	156.4	4.3 ± 0.1
184.8	4.7 ± 0.4	122.5	6.4 ± 2.7
225.4	10.1 ± 0.6	283.8	6.3 ± 1.8
777.5	6.3 ± 0.4	606.3	8.0 ± 1.0
837.8	6.1 ± 0.8	1031.0	7.0 ± 0.5
1598.5	7.4 ± 0.4	1642.8	9.1
lw	1.5 ± 1.3	lw	4.1 ± 0.5

lw - lake water DRP concentration before the stirring experiment.

± - indicates deviations from the mean derived from three parallel experiments.

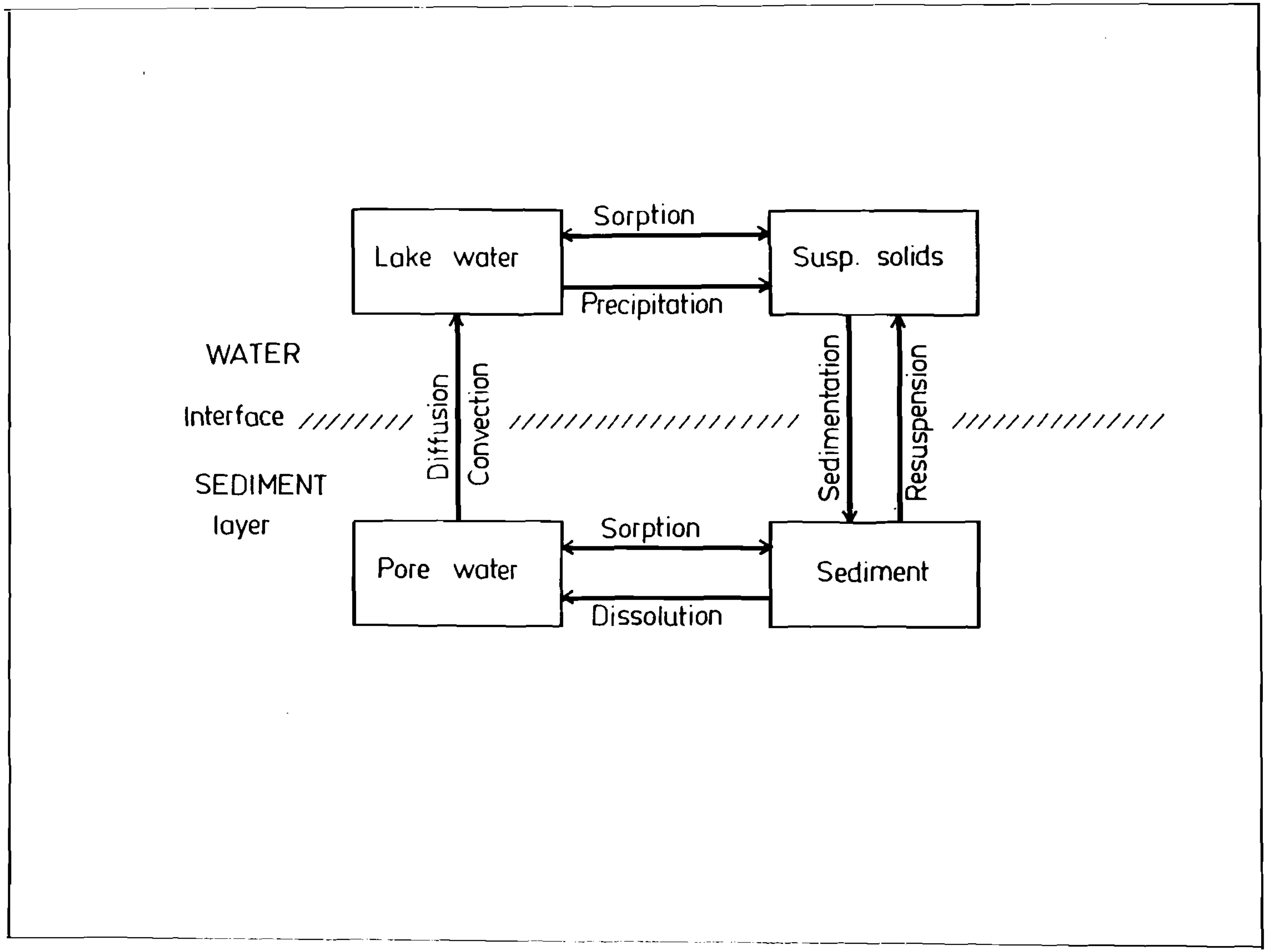

Figure 1. Major processes influencing the sediment-water interaction

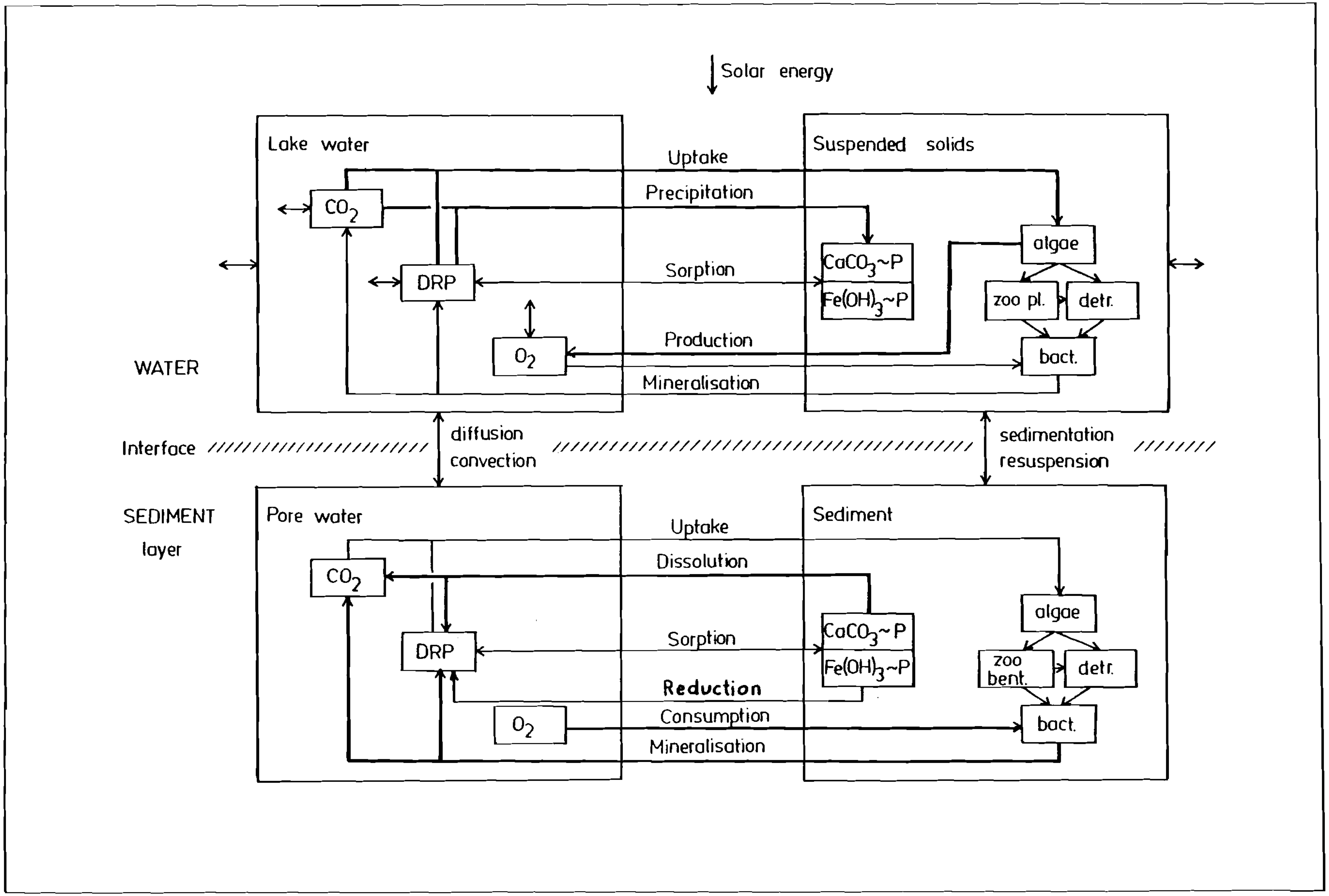

Figure 2. Main pathways of phosphorus cycle

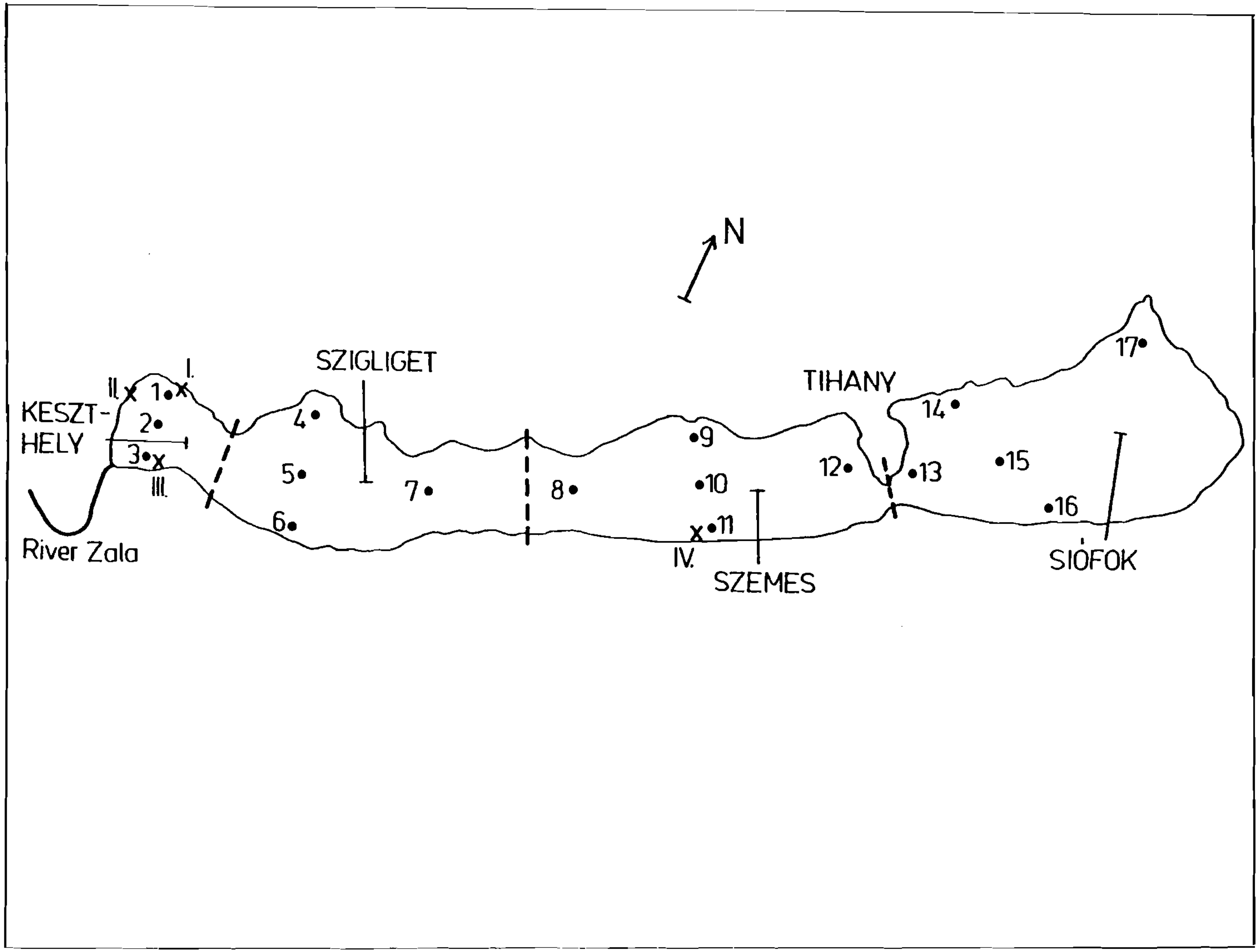

Figure 3. Sampling locations

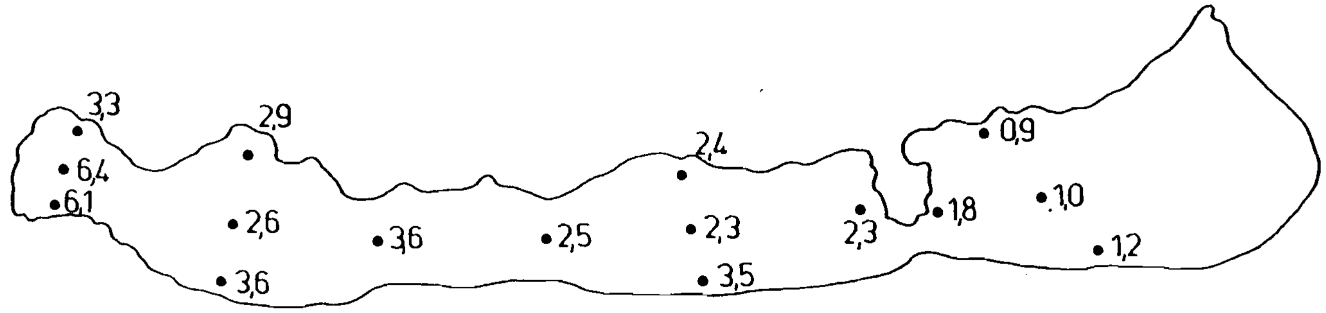

Figure 4. P:Ca ratio in the acid soluble fraction of the suspended solids (x 10^{-3})
6.04.1981

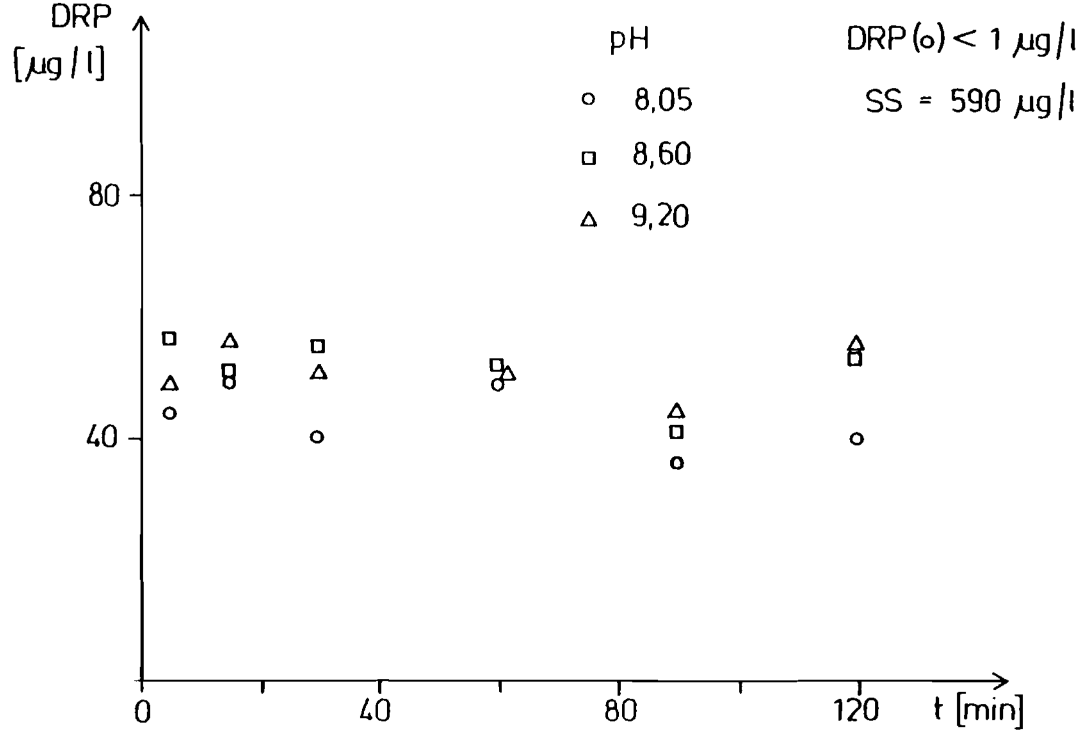

Figure 5. Time evolution of desorption at different pH values
Szemes sediment taken at 21.09.1981

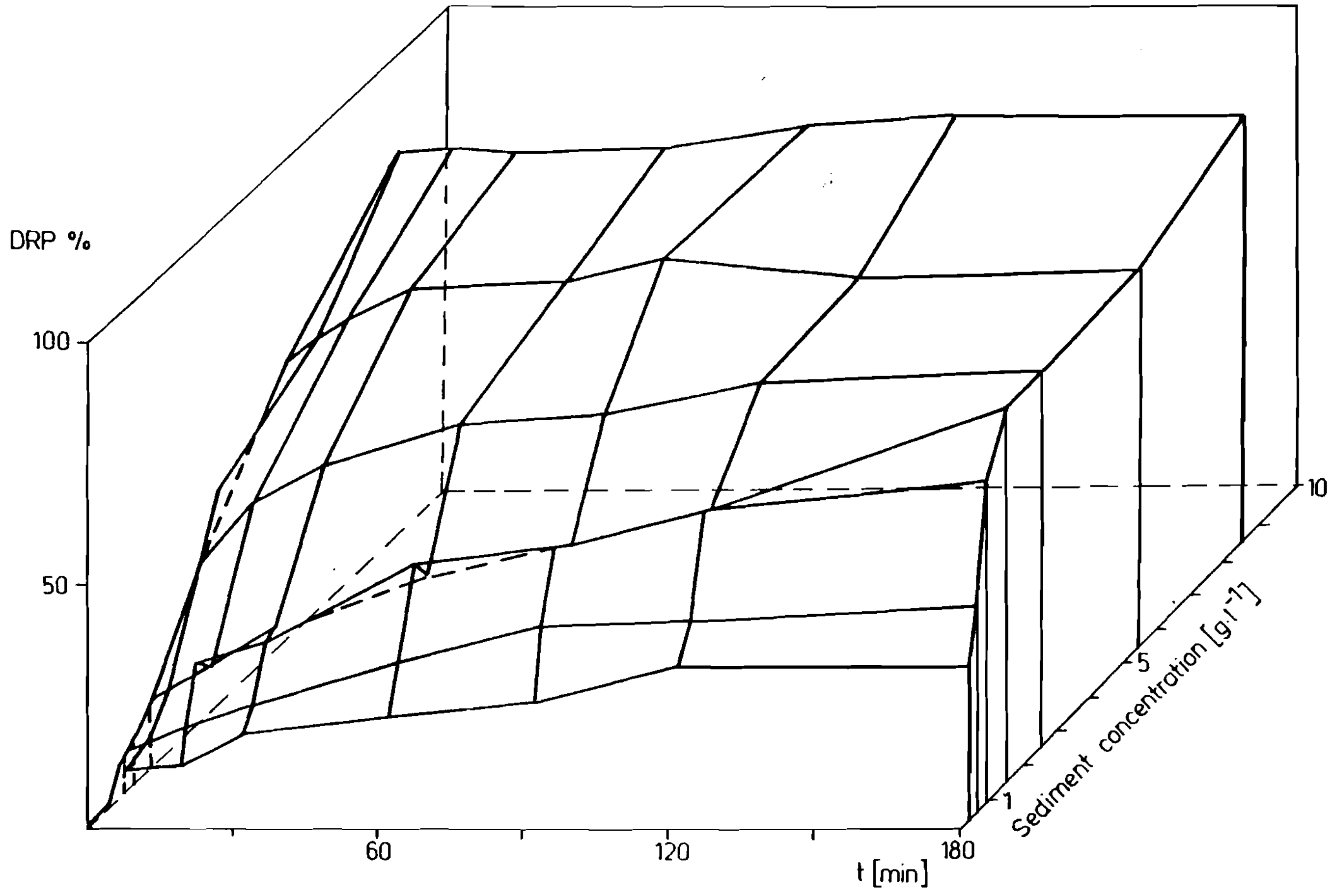

Figure 6. The change of the adsorbed DRP related to the initial concentration (Keszthely sediment)

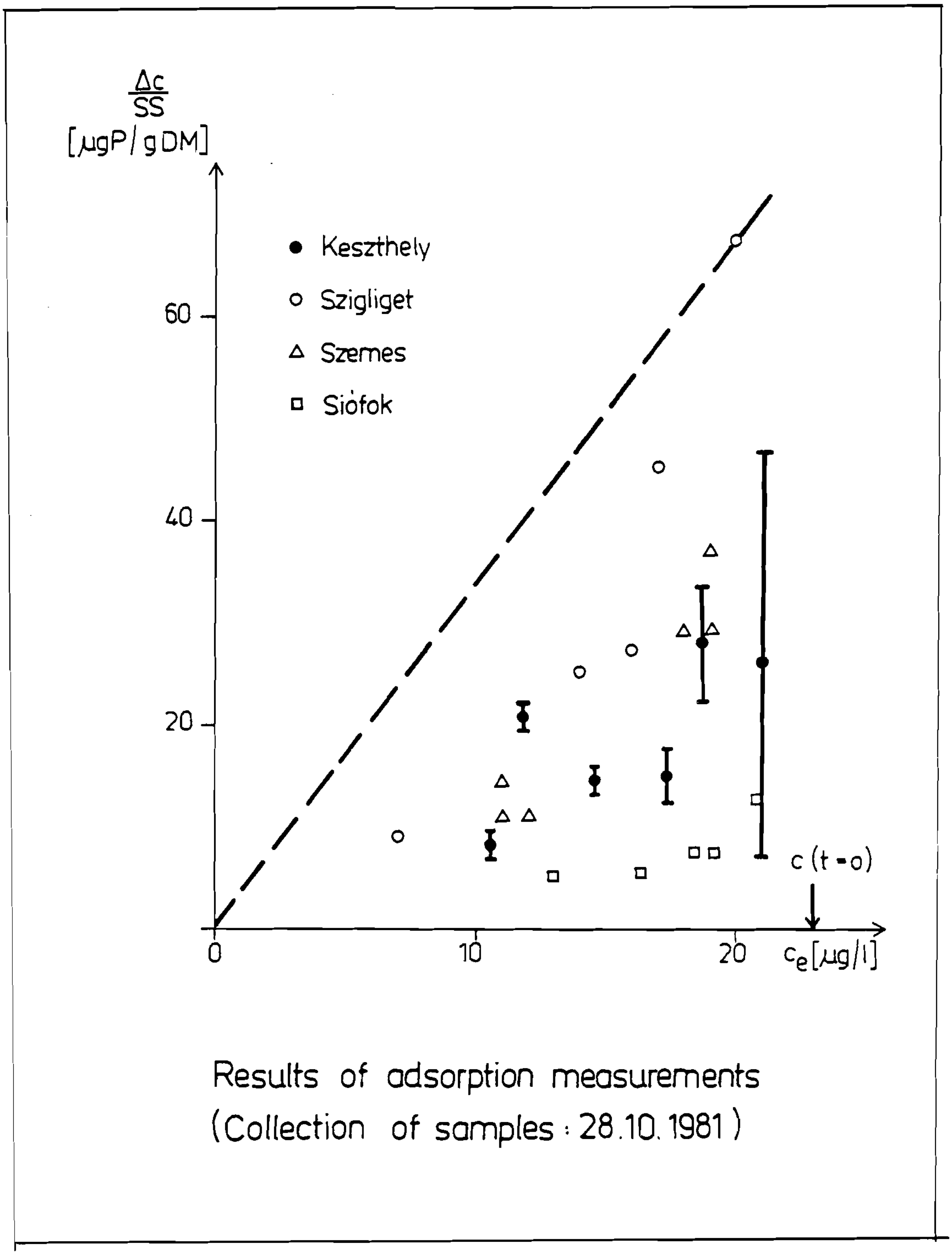

Figure 7. Results of adsorption measurements (collection of samples: 28.10.1981)

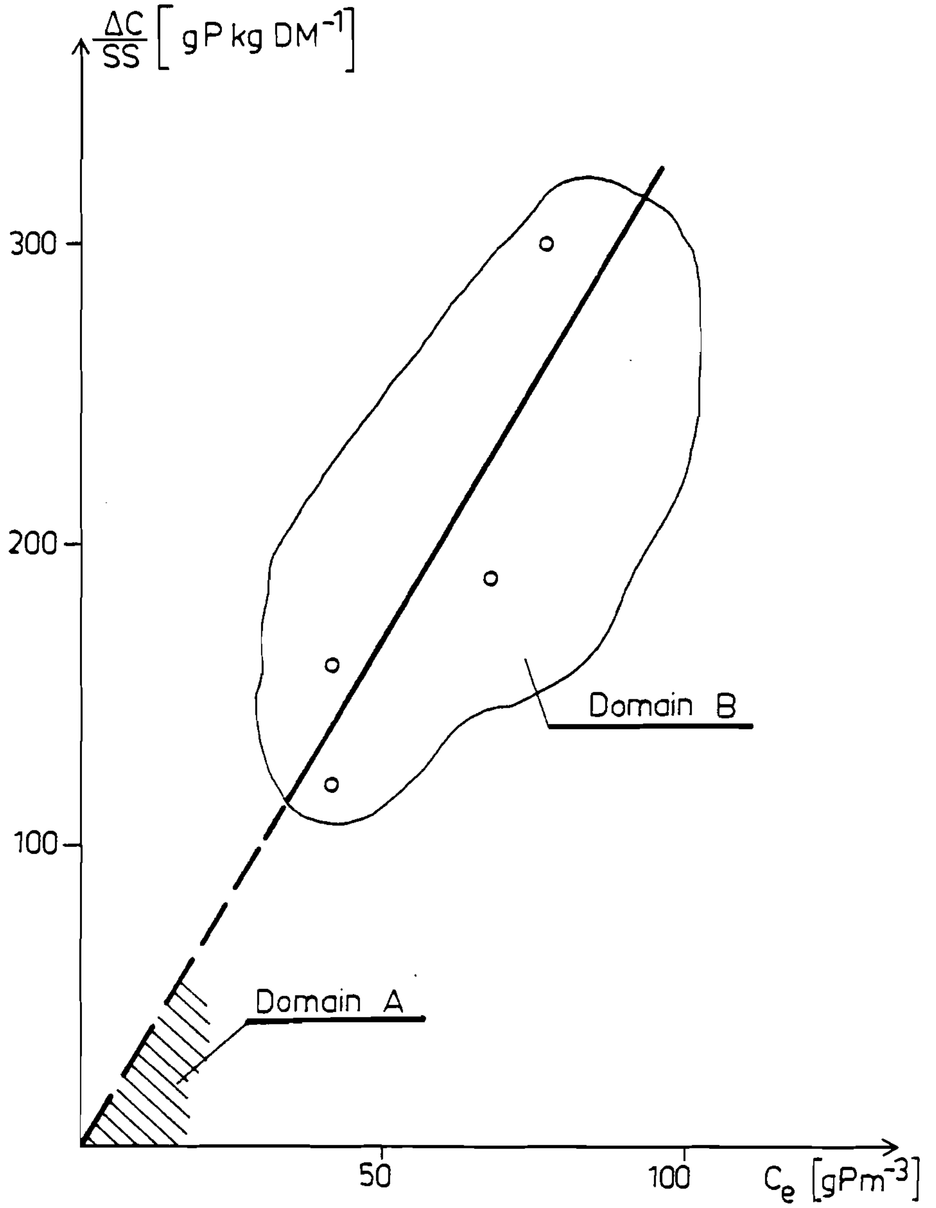

Figure 8. Domains covered by the two adsorption tests

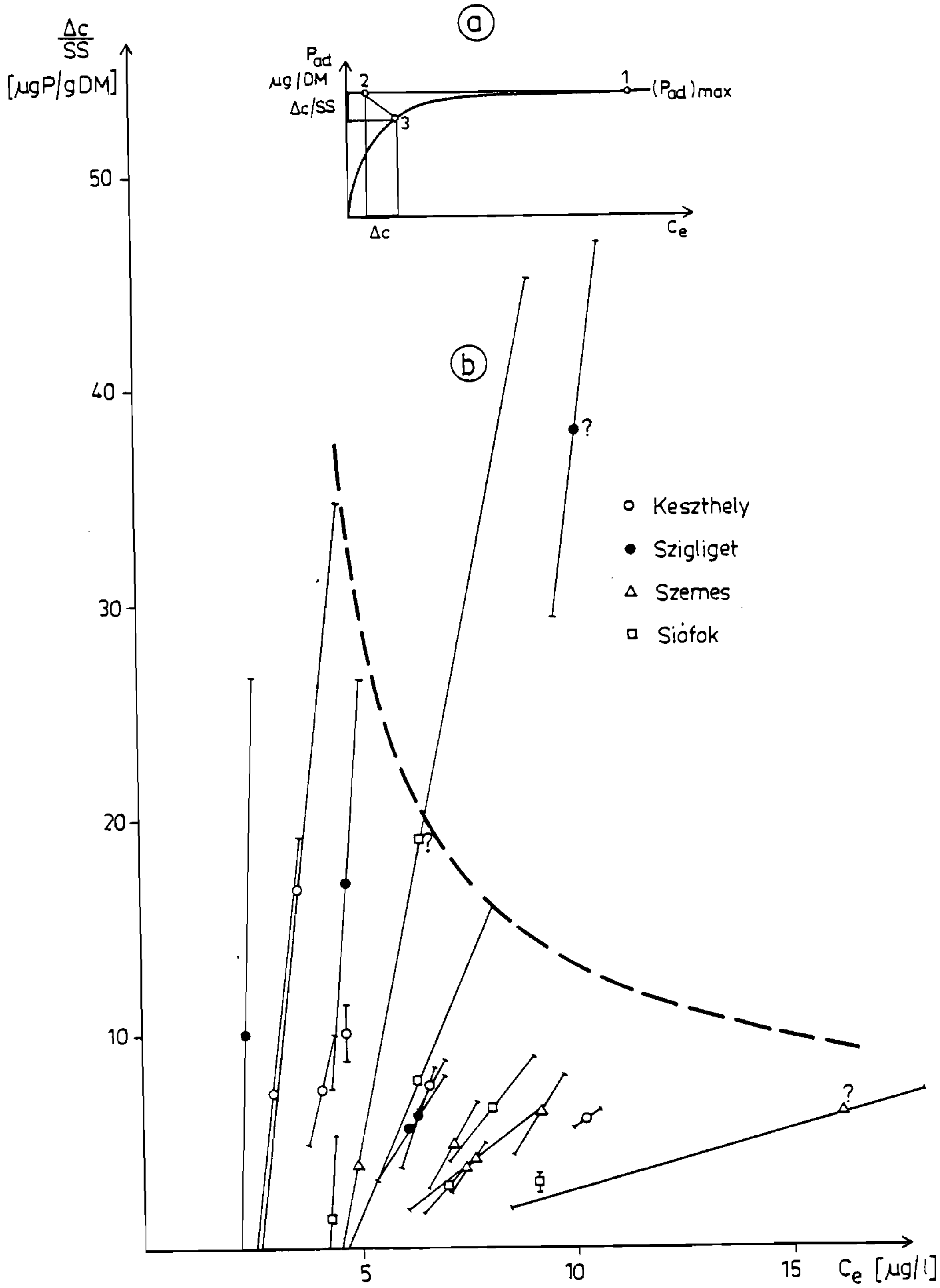

Figure 9. Results of the desorption experiment.
Sample collection: 28.10.1981

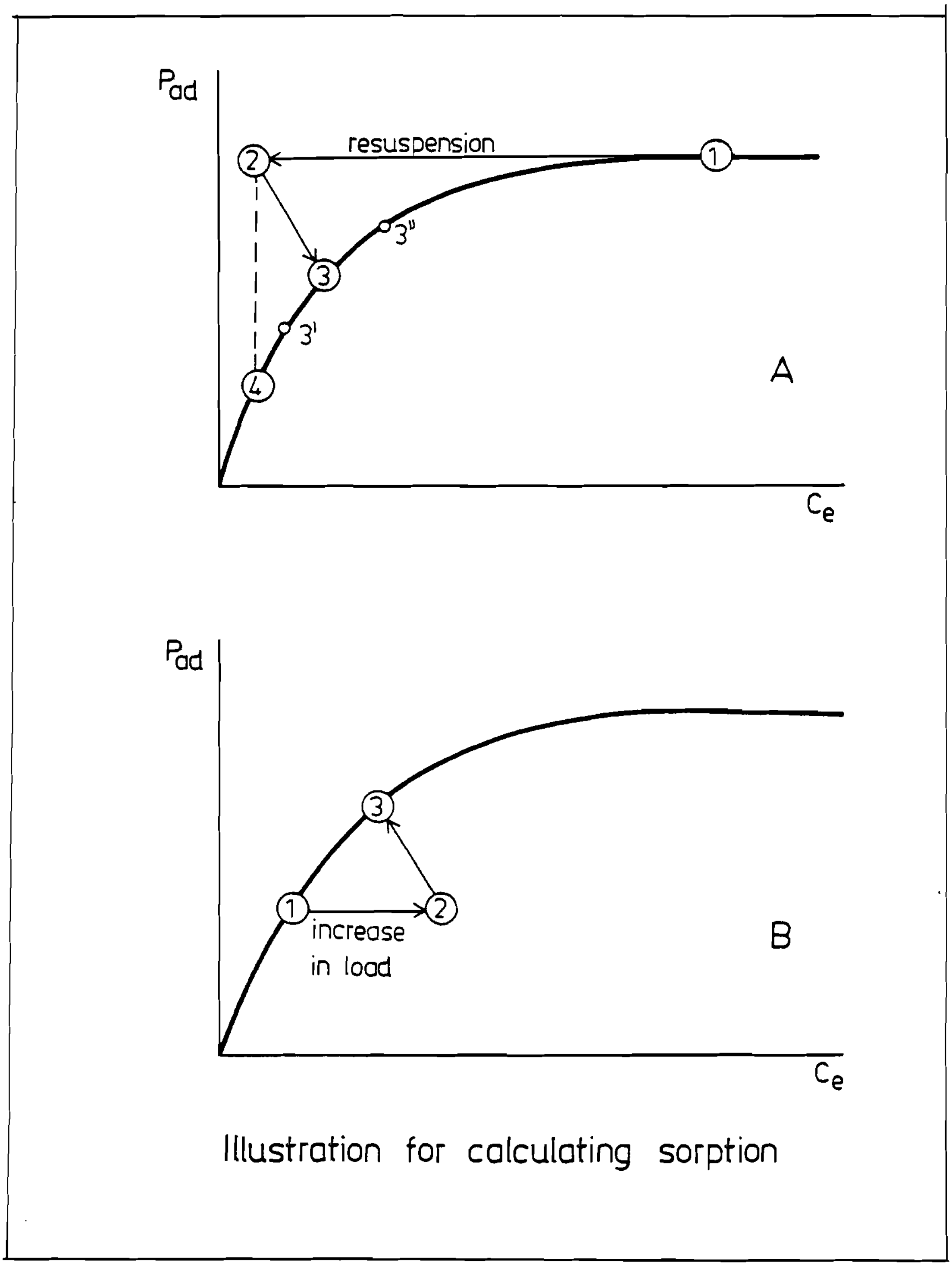

Figure 10. Illustration for calculating sorption

APPENDIX: Water quality of the pore water and lake water: daily observations of Szemes and Keszthely

Table 1. Summary of daily observations (meteorological factors and water quality parameters for the lake water and pore water, respectively) at Szemes, 20.05-03.06.1981

D A T E	Time of the sampling	Secchi depth (cm)	SS (mg/l)	Water Temperature (C°)	Wind velocity, daily average & approximate extremes (km/h)	Chl-a (µg/l)	Pheophytine (µg/l)	Conductivity, pore water (µs/cm)	Conductivity, lake water	pH conductivity, pore water	pH conductivity, lake water	diss. organic carbon, pw (mg/l)	diss. organic carbon, lw	NH4-N (mg/l)		PO4-P (mg/l)		Total dissolved P (mg/l)		Total dissolved iron (mg/l)	
														pw	lw	pw	lw			pw	lw
05.20	15.00	140	7.8	19.0	10.8±7.9	3.1	5.3	552±8	498	8.21±0.06	8.57	7.3±0.4	7.0	0.334±0.108	0.000	0.020±0.010	0.000	0.033±0.015	0.009	0.028±0.10	0.004
05.21	8.45	85	9.2	18.8	20.9±2.6	2.6	5.9	551±4	564	8.42±0.08	8.55	7.0±0.5	5.3	0.748±0.151	0.026	0.023±0.005	0.002	0.037±0.006	0.007	0.018±0.018	0.036
05.22	8.50		10.4	19.3	9.9±7.7	3.4	2.1	531.±19	534	8.08±0.07	8.58	7.3±0.5	6.0	0.343±0.183	0.020	0.012±0.008	0.000	0.063±0.022	0.014	0.004±0.004	0.000
05.23	8.45	58	30.7	18.4	24.5±5.0	3.1	5.1	582±5	578	7.78±0.14	8.52	7.0±0.4	8.0	0.303±0.189	0.011	0.017±0.012	0.005	0.055±0.023	0.018	0.000±0.000	0.000
05.24	7.00	57	14.7	19.6	5.1±2.7	2.8	3.8	572±3	465	7.92±0.04	8.53	7.7±0.8	7.5	0.513±0.055	0.010	0.041±0.007	0.010	-	-	0.037±0.025	0.000
05.25	7.00	70	-	20.4	5.4±5.5	2.0	3.5	588±13	586	7.98±0.05	8.56	6.8±0.2	7.0	0.544±0.111	0.020	0.011±0.004	0.000	0.018±0.003	0.006	0.016±0.032	0.000
05.26	7.00	55	9.8	20.2	7.2±4.8	4.0	3.6	621±12	615	8.05±0.09	8.62	6.7±0.4	8.0	0.413±0.078	0.024	0.025±0.004	0.004	0.056±0.011	0.014	0.021±0.028	0.000
05.27	6.15	50	20.5	20.3	6.1±4.2	7.0	7.1	600±27	622	8.51±0.01	8.62	6.8±0.9	7.0	0.312±0.062	0.006	0.019±0.007	0.003	0.039±0.010	0.009	0.002±0.004	0.000
05.28	7.00	58	13.3	18.8	7.6±3.3	5.1	2.7	573±8	600	8.16±0.28	8.42	6.5±0.3	6.6	0.522±0.140	0.047	0.049±0.020	0.003	0.050±0.020	0.010	0.032±0.023	0.246
05.29	7.00	57	31.6	19.3	15.1±2.2	1.7	8.7	567±7	578	7.91±0.07	8.52	7.1±0.4	6.6	0.516±0.124	0.000	0.034±0.012	0.000	0.037±0.018	0.008	0.132±0.101	0.000
06.01	7.00	75	8.8	19.0	11.3±5.5	2.3	2.9	587±13	586	8.47±0.03	8.54	7.3±0.4	6.8	0.750±0.133	0.027	0.058±0.017	0.002	0.066±0.011	0.009	0.045±0.038	0.055
06.02	8.00	147	2.7	22.4	7.8±8.2	2.8	3.4	596±34	622	8.30±0.15	-	8.1±0.2	8.5	0.748±0.172	0.009	0.024±0.008	0.000	0.060±0.010	0.012	0.021±0.004	0.009
06.03	7.00	171	4.9	24.2	3.9±4.3	2.0	2.3	631±10	694	8.50±0.17	8.55	7.7±1.3	7.0	0.644±0.147	0.000	0.026±0.009	0.001	0.050±0.012	0.029	0.043±0.014	0.008

Table 2: Summary of daily observations at Keszthely (4-14.08.1981)

KESZTHELY

Date	Secchi (cm)	SS (mg/l)	Tw (C°)	W* (m/s)	R (cal/m^{2}d)	Chl-a (μg/l)	pheophygment (μg/l)	pH	conductivity (μs/cm)	PO4-P (mg/l)	NH4-N (mg/l)	Tot. Fe (mg/l)
4.08.1981	42	15.2	26.0	7.1	223	53.7	19.8	pw 7.9±0.3 lw 8.9	577±55 465	0.35±0.18 0.02	4.1±3.2 0.13	0.17±0.1 0.02
6.08.1981	-	64.3	24.3	13.9	288	71.0	20.1	pw 8.1±0.1 lw 8.9	650±18 465	0.31±0.04 0.02	3.9±0.7 0	0 ±0 0
7.08.1981	30	63.4	24.3	9.9	280	67.9	21.5	pw 8.2±0.2 lw 9.1	577±63 460	0.33±0.12 0.02	2.9±1.5 0.02	0.07±0.05 0
8.08.1981	36	41.9	24.5	4.8	278	92.6	0.1	pw 8.1±0.2 lw 9.0	593±50 560	0.38±0.11 0.04	3.2±1.0 0	0.07±0.04 0
10.08.1981	-	-	25.6	8.8	225	-	-	pw 8.3±0.3 lw -	586±34 -	0.35±0.11 -	2.8±2.6 -	0.08±0.08 -
11.08.1981	-	13.9	24.6	14.8	215	51.8	55.5	pw 8.4±0.1 lw 8.7	648±95 505	0.34±0.10 0.01	1.3±0.2 0	0.07±0.002 0
12.08.1981	1	49.2	24.6	13.2	273	53.6	59.1	pw 8.3±0.1 lw 8.5	626±55 511	0.38±0.12 0.01	0.5±0.1 0.06	0.11±0.04 0.01
13.08.1981	36	37.2	24.5	9.5	201	53.4	71.6	pw 8.5±0.1 lw 9.1	654±58 515	0.26±0.05 0.01	3.3±5.2 0.04	0.06±0.06 0
14.08.1981	-	4.5	22.6	8.6	262	35.3	61.3	pw 8.5±0.1 lw 9.0	644±67 520	0.30±0.06 0.02	4.4±1.9 0.02	0.16±0.004 0.03

*Recorded at Szemes

± indicates σ from 5 data (2)

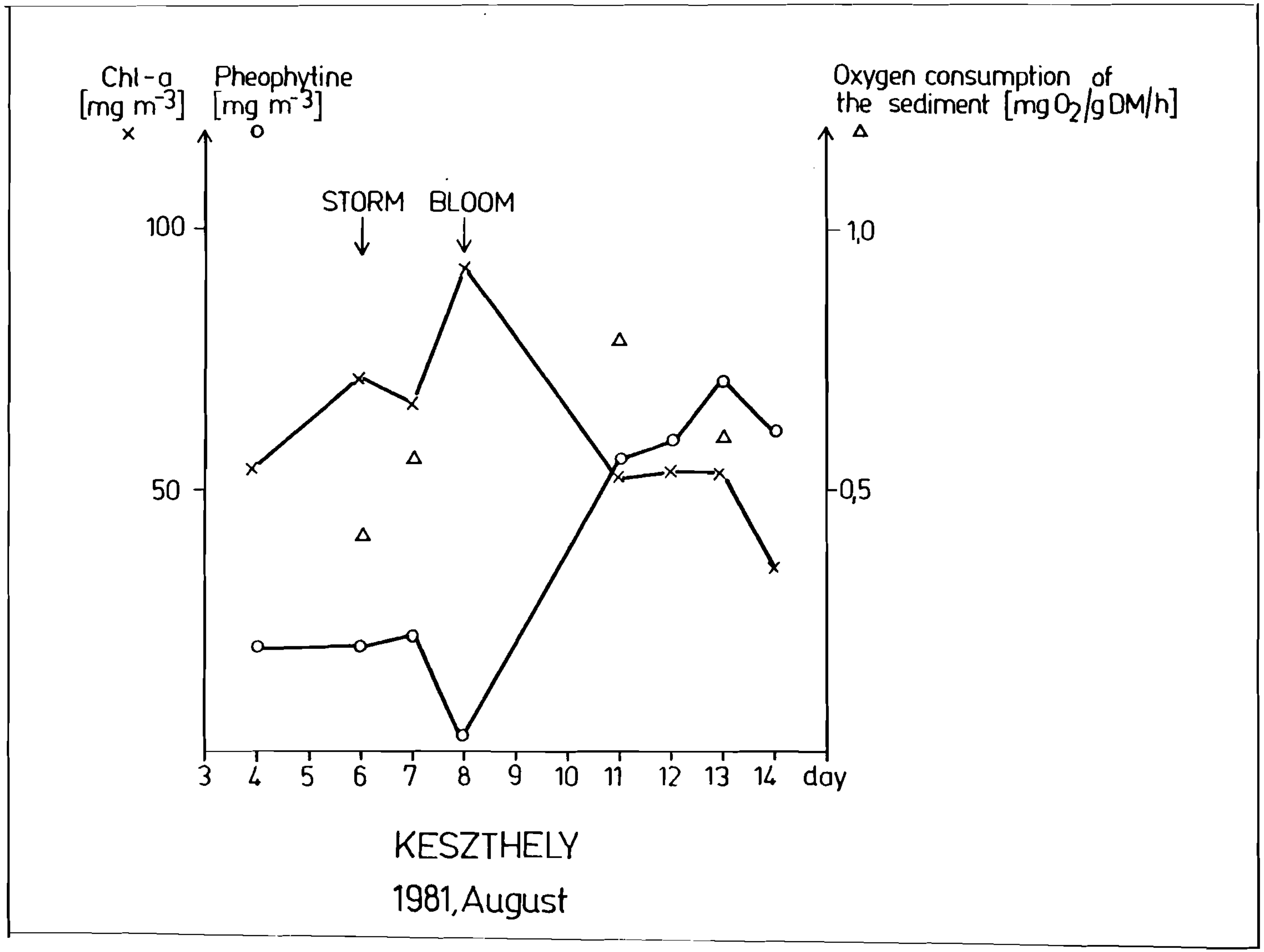

Figure 1. Change of Chl-a, pheophytine and oxygen consumption of the sediment
(3.08-14.08.1981)

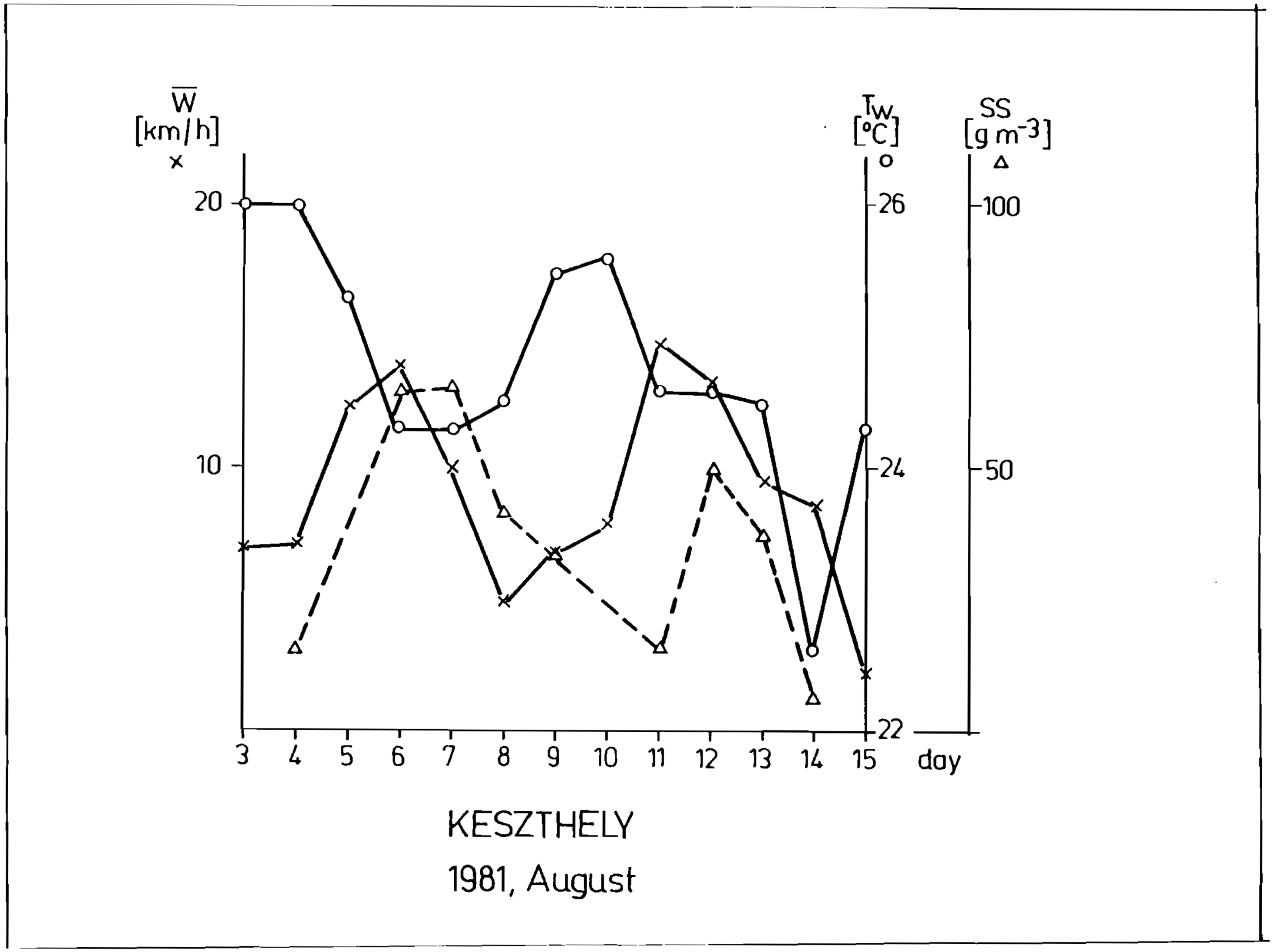

Figure 2. Change of meteorologic factors and the suspended solids (3.08-14.08.1981)

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