Displacement of Phosphorus in Structured Soils

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To my parents

Abstract

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Phosphorus losses from agriculture may enhance eutrophication of fresh water bodies. This thesis focuses on preferential flow as a phosphorus transport pathway. Both lysimeter and field plot observations were conducted to evaluate the significance of preferential flow for P losses and to test management practices to reduce P losses. A decision support system was also developed to identify critical source areas, to diagnose probable causes of P losses and to prescribe appropriate site-specific best management practices.

Preferential flow pathways are an important transport mechanism for P displacement from topsoil to drain tiles. Substantial drainage losses (on average 4.0 kg P ha^{-1}) of surface-applied P fertilizer, as measured in one of the studies included in this thesis, indicate a very effective vertical transport in structured clay soils. High sorption potential of the subsoil is bypassed because only a small part of the total pore volume is active in water/solute displacement.

Incorporation of P fertilizer and an increased contact between applied fertilizer and sorbing soil particles may significantly decrease P losses via preferential flow. Ponded flow conditions enhance P losses not only by enhancing preferential flow, but also by prolonging loading of P into water laying temporarily on the surface or subsurface.

During four years of observations, nine high discharge episodes were responsible for, more than a half of the total loads of suspended soil material and P losses from clay plots, although the water volume discharged during these episodes reached, only one fourth of the total discharge.

Different parts of the watershed do not contribute equally to total P loads. Identification of high-risk areas, site-specific diagnosis and abatement efforts should reduce the losses more efficiently. For instance, high-risk areas occupied no more than 5-10% of the total watershed area, according to decision support system developed in the study included in this thesis.

Research at different scales is necessary for better understanding of the P problem, as well as for an easier practical application of research results. Shifting the scales tests the relevancy of the research and helps the identification of the gaps in our knowledge and understanding of the problem.

Key words: preferential flow, leaching, lysimeters, field plot, watershed, P index, ponded flow conditions

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Contents

Preface

Papers I-IV

This thesis is based on the following papers, which are referred to in the text by their Roman numerals:

I. Djodjic, F., Bergström L., Ulén B. & Shirmohammadi A. 1999. Mode of transport of surface-applied phosphorus-33 through a clay and a sandy soil. *Journal of Environmental Quality* 28, 1273-1282.

II. Djodjic, F., Bergström L. & Ulén B. Phosphorus losses from a structured clay soil in relation to tillage practices. (Manuscript submitted to *Soil Use and Management*).

III. Djodjic, F., Ulén B. & Bergström L. 2000. Temporal and spatial variations of phosphorus losses and drainage in a structured clay soil. *Water research* 34, 1687-1695.

IV. Djodjic, F., Montas H., Shirmohammadi A., Bergström L. & Ulén B. A decision support system for phosphorus management at a watershed scale. (Manuscript submitted to *Journal of Environmental Quality*).

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Introduction

Bringing light…

The story of phosphorus (P) begins in 1699 when the German chemist Henning Brand, who was seeking for a way to transform base metals to gold, discovered a white, waxy solid. Henning Brand isolated P from putrid urine, but today the sources of P are phosphate mineral rocks known as apatites ($Ca_{10}[X_2PO_4]_6$, where X= OH^- or F^-, and Ca^{2+} may be substituted with Na^- or Mg^{2+}). Phosphorus is a non-metal belonging to the Group 5A of the periodic table of elements, and its common physical form is a wax-like white solid (white P) or red solid (red P). The solid has a relatively high vapor pressure and the reaction between phosphorus vapor and oxygen in air results in phosphorescent glow. This glow gave the element its name – *phos* in Greek means light, and *phorus* means bringing. However, elemental P does not have much use and it is usually reoxidized to P_2O_5, or rather P_4O_{10}, because the structure of each P oxide is based on P tetrahedron (Petrucci & Harwood, 1997). In contrast, phosphorus compounds are widely used. For instance, phosphoric acid (H_3PO_4) is used in production of baking powders and instant cereals, in cheese making and in soft drinks. Phosphine (PH_3) is poisonous and has been used in pesticides. Phosphorus trichloride (PCl_3) is used in production of a vast range of products: soaps and detergents, plastics and synthetic rubber, motor oils and pesticides. The polyphosphates and metaphosphates are used in detergents where they act as water softeners by complexing with metal ions in the water.

Compounds of P are also of vital importance to living organisms. Phosphate groups are present in both deoxyribonucleic acids (DNA), in which information about inherent traits resides, and adenosine triphosphate (ATP), the energy-containing molecule of living organisms. Plant roots absorb P mainly in soluble inorganic form (phosphate anions) and, to a lesser extent, in the form of organic compounds (Brady, 1990). The addition of P has long been recognized as necessary to increase production in modern agriculture. Phosphorus fertilizers, such as superphosphate, $Ca(H_2PO_4)_2{\cdot}H_2O$, are produced from the above-mentioned apatites. Approximately 90% of mined apatites are used for fertilizer production. For instance, triple superphosphate, which contains 20 to 21% P, is produced in a two-step process where phosphoric acid is produced by the action of sulfuric acid on phosphate rock and then the phosphate rock is treated with the concentrated phosphoric acid produced in the first step. Phosphorus sources are quite large, but limited and unevenly distributed around the world. The estimations of the longevity of P reserves vary from 85 to 400 years, depending on the prognosis for P fertilizer consumption and the assumed profitability of phosphate mining. Due to the low prices of P fertilizers between the 1950s and 1990s, and their positive effect on crop production, an increased usage of P

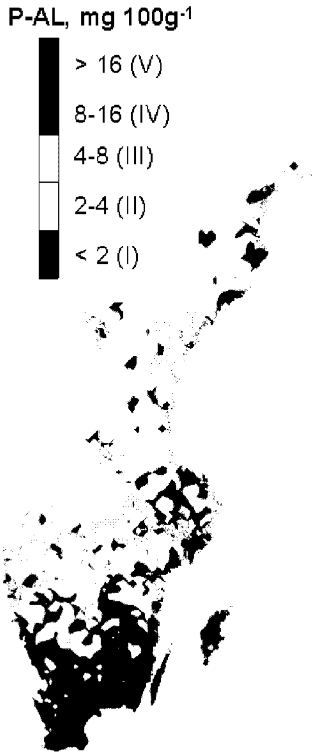

Fig. 1. Ammonium lactate-acetate-soluble phosphorus (P-AL) in the plow layer. Classes on the map are those used when rating soil fertility. From Eriksson *et al.* (1997).

fertilizers resulted in accumulation of P in agricultural soils in most of the western countries. In Sweden, a build-up of P reserves in agricultural soil during a 40-year period is estimated at 600-700 kg ha^{-1} (Andersson *et al.,* 2000). Furthermore, Eriksson *et al.* (1997, Fig. 1) concluded that only 13.5 percent out of totally 3100 Swedish arable soil samples belonged to the two lowest P classes, according to the five-class determination system based on the ammonium lactate soluble P determination (P-AL) (Egner *et al.,* 1960).

The other main source of P in modern agriculture is manure. As a necessary component of the nucleic acids and nucleotides, and essential for the formation of bones and teeth, P is a common addition to animal feeds in modern cattle management. However, the utilization of P from the animal feeds is quite low, especially in intensive pig and poultry systems. For example, P retention was only 35 and 14% of the P intake for hogs and sows, respectively (Poulsen, 2000). This, in combination with the high densities of animals in certain geographical locations, produces large quantities of P that exceed the demands of arable farming. In Sweden, animal production is concentrated in the southern part of the country, especially in the counties of Halland, Småland, Skåne and Blekinge.

...or extinguishing it?

As one of the major nutrients, P may enhance the eutrophication processes in the receiving water bodies (Schindler, 1977). Eutrophication is a natural aging process of lakes and streams caused by nutrient enrichment. Human activities, such as agriculture and industry, can rapidly accelerate nutrient inputs and thereby eutrophication itself. The increased growth of undesired algal populations followed by the depletion of the oxygen caused by algal death and decomposition restricts the use of freshwater reservoirs. The species composition shifts and the nutrient status of the water recipient is altered to a higher nutrient level. For instance, in Sweden, 14 000 out of a total of 95 700 lakes larger than one hectare have total P concentrations exceeding 25 $\mu g\ l^{-1}$ and therefore are classed as eutrophic. Furthermore, 81 lakes are classed as hypereutrophic, with total P concentrations exceeding 100 $\mu g l^{-1}$ (http://info1.ma.slu.se/Miljotillst/Eutrofiering /Hypertrofikarta .ssi, Accessed 20 March 2001). The areas with eutrophic lakes in Sweden are shown in Fig. 2.

The first and probably the easiest step in the abatement of the eutrophication problem was to reduce the point P sources, such as waste from urban areas and industry. The efficiency of P removal from sewage is very high. In the processing of sewage, polyphosphates are degraded to orthophosphates by bacterial action.

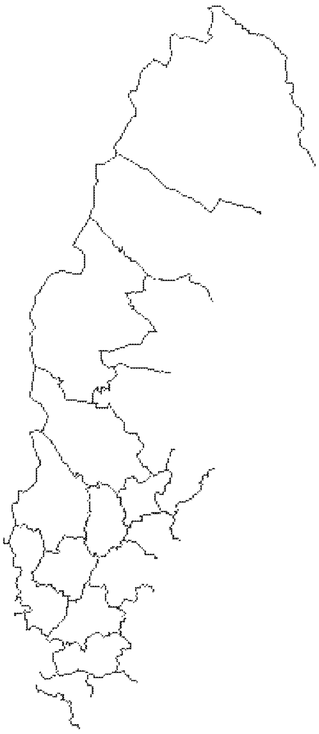

Fig. 2. Areas (colored green) with eutrophic lakes in Sweden (http://www.environ.se/ index.php3?main=/dokument/fororen/overgod/eutro/omr.html: Accessed 20 March 2001).

The orthophosphates may then be precipitated either as iron, aluminum or calcium phosphates or as hydroxyapatite. The precipitating agents are generally aluminum sulfate, iron chloride or calcium hydroxide. In a modern sewage treatment plant, up to 98% of the phosphates in sewage can be removed. According to the Swedish Environmental Protection Agency, 95% of the waste from municipalities is chemically and biologically treated in Sweden.

The reduction of point-source contributions to eutrophication is, however, not sufficient for sustainable management of impaired freshwater ecosystems. A more holistic approach is required, where non-point P sources within a given watershed and affected water body are linked together and treated as one unit. Recently, agriculture as a source of P inputs to surface water has gained increased attention from water-resource managers. The estimations of P contributions to water bodies from agriculture vary. For instance, according to Löfgren & Olsson (1990) the agricultural contribution of P to surface waters in Sweden was, on average, 621 t P yr^{-1}. Larsson (1997) estimated that the agricultural contribution to the total P loading to surface waters in Sweden is about 900 t P yr^{-1}, *i.e.* agriculture was the largest single contributor to P loads (Fig. 3).

The diffuse character of P losses from agriculture, in combination with the complex behavior of P in the environment, makes the abatement work much more difficult than in the case of finding countermeasures to losses from point sources. Processes and factors affecting P behavior in soil at different scales (soil sample, soil profile, field, watershed) are still not fully known. Especially the quantification of different physical, chemical and biological processes of

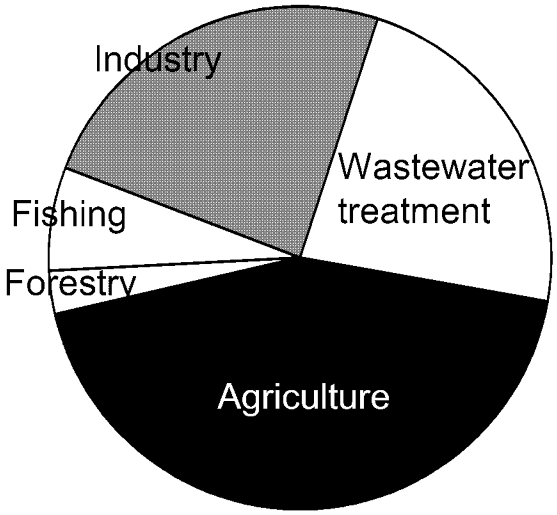

Fig. 3. Sources of P loads to surface waters in Sweden. Adapted from Larsson (1997). The losses from private households, which are not connected to wastewater treatment plants, are not included.

importance for P behavior in the environment is a great challenge for all parties (i.*e.* researchers, farmers and decision-makers) involved in agricultural production and environmental protection. Therefore, the line separating P as an asset in agriculture from P as an environmental risk is still very blurred. To make it more distinct we have to be able to identify and quantify the most important factors for P behavior, and to account for their temporal and spatial variations at different scales.

Phosphorus in soil

Soil P exists in both inorganic and organic forms. The distribution of these two main forms of P primarily depends on soil properties. In general, the organic forms range from 30 to 65% (Harrison, 1987). Labile P in the soil solution is a very small part of the total P content in soil and its concentration ranges between 0.01 and 3.0 mg P l^{-1} (Frossard *et al.*, 2000). These concentrations are not sufficient to fulfill plant requirements, and the remaining part must be obtained through a set of chemical and biological processes which release/retain P from the other pools. The most important abiotic processes are dissolution-precipitation and sorption-desorption, whereas the most important biological process is mineralization-immobilization.

Dissolution of apatite, the most common phosphate mineral in the soil crust, is induced by the action of H^+ ions, which may originate from soil itself, from plant roots or microbes. This process is also known as weathering and it is the main source of nutrients in natural, undisturbed soils. Precipitation of P in soil depends on the soil properties, mainly pH and the presence of either Ca carbonates or Al/Fe hydrous oxides. In calcareous soils, P is precipitated in a range of various Ca phosphates, although seldom as apatite due to the presence of Mg and organic compounds, which inhibit the crystal growth of apatite. In Al and Fe systems, P is precipitated through the formation of amorphous or crystalline Al and Fe phosphates. Precipitation-dissolution processes are governed by the solubility of the least soluble product involved in the reactions.

Sorption-desorption processes are governed by the concentrations of phosphate anions or organic P in the soil solution. The term sorption here includes both the rapid surface reaction of adsorption and the long-term reaction of absorption into porous material. The most important minerals in soil regarding P sorption and

Table 1. *Phosphorus pools in soil, relative size and availability*

	Dissolved P ↔ Fast process	Reversibly sorbed P ↔ Slow process	Stabile pool
Location	In soil solution	On oxide surfaces	Mineral lattices, organic matter, occluded P
Reserves in soil	Small reserves	Amount varies	Large reserves
Relative size	1	100	5000
Availability	Immediately available	Available	Practically unavailable

desorption are Al and Fe oxides, hydroxides and hydrated oxides, Ca carbonates and, to lesser extent, clay minerals. For instance, in neutral and alkaline soils, P is precipitated as the sparingly soluble $Ca_3(PO_4)_2$. Oxides or hydroxides of Al and Fe may be positively, negatively or zero charged. Phosphate adsorption usually occurs as specific adsorption via ligand exchange on positively charged oxides or hydroxides. OH^- or H_2O is released and a phosphate surface complex is formed. Eventually, Al and Fe oxide and hydrated oxide coatings can surround the adsorbed P, decreasing its availability to further transformations. This process is known as aging or occlusion. Clay minerals sorb less P than oxides and the sorption takes place at the edges of the octahedric layer of clay minerals. These broken edges behave similarly regarding P sorption as the Al hydroxide surfaces.

Phosphorus uptake by microorganisms and higher plants and the following decomposition and mineralization of dead microorganisms, plant material, and litter are important parts of the P cycle in soil. The most resistant P organic compounds in soil are monoesters such as inositol-phosphate whereas other organic P compounds (such as nucleic acids and phospholipids) are held less strongly by the soil matrix and therefore more easily transformed to inorganic P (Magid *et al.,* 1996).

The complexity of P forms, processes and factors involved in P cycling creates an enormous challenge when attempting to properly and accurately describe P fluxes and P pools in soil. From an agronomic point of view, soil P tests (such as P-AL, Olsen P, Mehlich 1, Mehlich 3 *etc.*) were designed and applied to extract and quantify the soluble and easily desorbed P as an indicator of P amounts available for crops during the growing season. The agronomic soil P testing is often the only available information about P status in a certain soil, and therefore soil P testing is even used to asses the environmental risk of a certain site. Some results show positive correlation between the agronomic soil test P and P losses in run-off and drainage (Heckrath *et al.*, 1995; Pote *et al.*, 1996; Sibbesen & Sharpley, 1997; Beauchemin *et al.*, 1998; Sims, 1998; Hesketh & Brookes, 2000; Pautler & Sims, 2000). However, it is difficult to make a general correlation due to the influence of site-specific factors, such as P transport characteristics, climatic, and hydrologic site properties. Heckrath *et al.* (1995) identified a "Change–Point" for a soil at Broadbalk, UK (57 mg Olsen P kg^{-1}), after which further increase in soil P test value resulted in high P losses. This "Change–Point" value is quite high, and significant P losses have occurred from soils with much lower soil P testing values. Hesketh & Brookes (2000) used 0.01M $CaCl_2$ to estimate the "Change–Point" and found that it varied for different soils between 10 and 119 mg Olsen P kg^{-1} soil. The lack of a single standard method for soil P analyses is another problem, which complicates the comparison of values obtained with different soil P testing methods around the world. However, soil P test measurements are extremely valuable. New technologies and software development (*e.g.* Geographic Information System (GIS)) may help us to better account for the spatial and temporal variations in soil P testing values, as well as to combine this new information with other spatially and temporally varying information, such as soil properties, topography, hydrology *etc.*, to better asses the environmental vulnerability of a certain site.

Phosphorus transport

Phosphorus transport from surrounding fields to a water recipient occurs in two main forms: in dissolved and in particulate form. The particulate form of P includes both P sorbed on soil particles as well as P included in organic compounds. The distribution between dissolved and particulate P forms varies over a wide range, depending on the main transport pathway, soil properties and management practices, and hydrological features of the specific site. It is important to stress that the dissolved P is immediately available for algae, whereas the availability of particulate P varies due to the choice of the method for bioavailability determination, and site-specific properties such as soil type and agricultural management practices. For instance, in Scandinavia, the availability of particulate P varied between 5 and 41% depending on the above-mentioned factors (Rekolainen *et al.*, 1997).

The main transport pathways for P removal from agricultural soils are surface run-off (overland flow), interflow (lateral flow below the soil surface), matrix flow and preferential flow (Haygarth & Sharpley, 2000). In many cases, there is no distinct separation between different pathway terms and all of them may be active in P displacement simultaneously. However, in most cases, one of the pathways is dominant and its identification is necessary for potential site-specific prevention and abatement of P losses. Site-specific P sources and P transport conditions should be the basis for the choice of site-specific Best Management Practices (BMP).

The evidence of P losses via surface run-off and interflow is well documented (Svendsen & Kronvang, 1991; Sharpley *et al.*, 1994; Catt *et al.*, 1998). Both dissolved and particulate P are transported via these pathways and their distribution depends on the present land use and management of the upstream fields. In general, particulate P dominates in surface run-off from cultivated fields, whereas the overland flow from grass- and forestland mobilizes little sediment and, therefore, the particulate P contribution to the total P losses decreases. Surface run-off driven P losses are closely connected with the erosion issue, and erosion countermeasures are also often effective for P loss reduction. The amount of transported P depends on soil properties (infiltration capacity, soil P content, soil texture and erodibility), site characteristics (slope, vegetation cover and roughness coefficient) and precipitation duration and intensity.

Matrix and preferential flow pathways refer to vertical water and solute transport. Matrix flow stands for uniform water movement through the whole pore volume of a soil profile, whereas preferential flow of water occurs along preferential pathways such as macropores, cracks, and wormholes (Beven & Germann, 1982) where only a small part of the total pore volume is involved in water and solute displacement. Phosphorus export via matrix and preferential flow pathways is less well documented but has gained increased attention during the last couple of years. Phosphorus concentrations in percolating water are assumed to be small, due to the high sorption potential of P-deficient subsoils. However, high P concentrations measured in drainage water in recent studies forced a more detailed approach to this issue, especially in tile-drained fields where overland flow was reduced, but the P export problem remained. For instance, Sharpley & Rekolainen (1997) identified several situations where soils are more vulnerable to P export through the soil profile: i) soils with a low P sorption capacity (acid organic and peaty soils, and sandy soils), ii) waterlogged soils where the conversion of Fe(III) to Fe(II) leads to P release, or organic P mineralization, and iii) soils with preferential flow through macropores and earthworm holes. Even in the case of flow along preferential pathways, the distribution of P forms varies. For instance, Heckrath *et al.* (1995) and Stamm *et al.* (1998) found that dissolved P dominated P leaching losses, whereas Bauchemin *et al.* (1998) and Ulén *et al.* (1998) identified particulate P as the major P form in water effluent. Simard *et al.* (2000) concluded that P concentrations and forms in drainage water were influenced by soil texture, tillage intensity and frequency, cropping systems and seasonal variability.

As mentioned above, 14 000 lakes in Sweden have a eutrophic status. Clay soils are the dominant soil type in a large number of the watersheds that are drained to these eutrophic and hypereutrophic lakes (Figs. 2 and 4). According to Eriksson *et al.* (1999) (Fig. 4), 55% of agricultural soils in Sweden have a clay percentage exceeding 15%.

Over 1.2 million hectares of a total of 2.75 million hectares of arable land in Sweden are tile drained (SCB, 1984; SCB, 2000), including the majority of clay soils. High P losses in drainage discharge caused by preferential (macropore) flow have been recorded from fields and catchments with clay content varying over a wide range, from 15 to over 60% (Gaynor & Findlay, 1995; Turtola & Jaakkola, 1995; Stamm *et al.*, 1998; Hooda *et al.* 1999; Laubel *et al.*, 1999). If we assume that preferential flow is a shortcut for P transport (either soil P or fertilizer/manure derived P) vertically through the soil profile, and that the efficient drainage transport through tile-drains enables rapid lateral P transport from tile-drained fields to water recipients, the combination of these two conditions creates an undesired, but highly efficient pathway for P displacement from the topsoil into adjacent freshwater bodies.

This hypothesis was the starting point for the work that I have been involved in during the last four years, which resulted in this thesis. Along the way, hypotheses were tested in a lysimeter study, field observations were made and analyzed, and different prevention methods that were assumed to minimize P losses were tested.

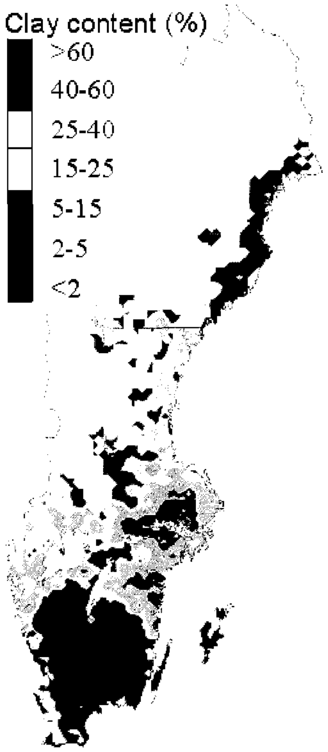

Fig. 4. Clay contents in agricultural topsoils (0-20 cm). From Eriksson *et al.* (1999).

Finally, an attempt was made to assess the issue of P loss as a whole with its spatial and temporal variations, as well as to develop a method for site-specific risk assessment and abatement. The width and depth of the eutrophication issue in general and P losses from agriculture in particular made this journey very interesting and enjoyable.

Objectives of the work included in this thesis

The overall aim of the work included in this thesis was to evaluate the significance of fertilizer P as a source and preferential flow as a transport pathway for P losses from structured clay soils. The specific objectives of the different studies included in this thesis were:

- evaluation of the significance of preferential flow pathways for P transport through the soil profile;
- evaluation of the dominating P form in percolating water;
- determination of the P source (soil P or fertilizer P);
- assessment of the impact of different management systems on P losses through the soil profile;

- estimation of the impact of flow rates and high discharge episodes on P leaching at field scale;
- investigation of the relationship between concentrations of suspended material and particulate P;
- identification of areas vulnerable for P losses at watershed scale; and
- site specific abatement of P losses based on site-specific source and transport characteristics.

Material and methods

How were the studies carried out?

Undisturbed soil monoliths (0.295 m inner diameter and 1.18 m in length, Fig. 5) were used for evaluation of the significance of preferential flow for P transport (Paper I), and for the assessment of different abatement measures (Paper II). The study described in Paper I was carried out during four weeks under controlled environmental conditions. An equivalent of 100 kg P ha^{-1} labeled with the radioactive isotope ^{33}P was applied on the soil surface of five clay (Lanna) and three sand (Mellby) monoliths. The following day, and then successively after 1, 2, 3 and 4 weeks, 100 mm of water (in total 500 mm) was added under ponded conditions to induce leaching. All leachate was collected and analyzed for total P and dissolved P, as well as for the ^{33}P content.

In total 13 Lanna clay monoliths, which were exposed to three different treatments, were used in the study described in Paper II: five monoliths in “no-till treatment”, where P fertilizer was added to the soil surface without any management practices; five monoliths in “conventional tillage treatment”, where preferential pathways were destroyed by tillage before surface application of P fertilizer; and three monoliths in “P fertilizer incorporation treatment”, where soil and P fertilizer were mixed after application. All soil monoliths were exposed to natural climatic conditions from July 1998 to May 2001. Barley was grown in all lysimeters during the vegetation seasons. Phosphorus fertilizer labeled with radioactive isotope ^{33}P (100 kg P ha^{-1}) was applied on two occasions with a 2-yr interval between the applications. All leachate was collected and analyzed for total P, dissolved P, and ^{33}P concentrations.

The results obtained in the lysimeter studies should be viewed with caution when evaluating their relevance for field conditions. The plot observations from the Lanna site analyzed in Paper III were therefore very helpful as a comparison since they better reflect field conditions. Water discharge, total P and dissolved P concentrations, and concentrations of suspended material from four field plots (0.4 ha) at the Lanna site in south-west Sweden were measured during a four-year period (1993-1997).

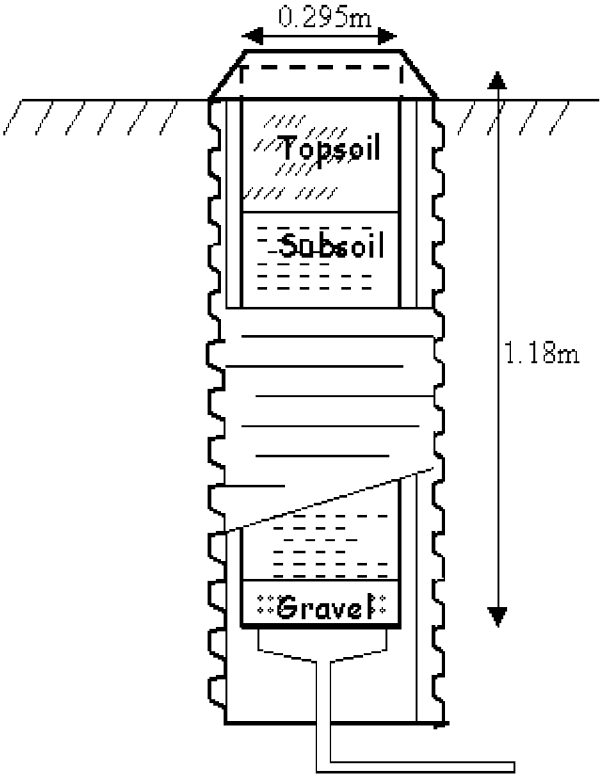

Fig. 5. Lysimeter placed in a belowground pipe. From Bergström & Johansson (1991).

Finally, the development of the Decision Support System (DSS) in Paper IV offered an opportunity to study the P issue with a more holistic approach, but also to evaluate the importance of the work that had been done in the previous studies (Papers I-III). A quite detailed data set representing soil properties, topography, land use and management in a small, agricultural watershed in south-western Sweden (Vemmenhög) was evaluated and processed with the raster-based GIS software ERDAS IMAGINE 8.4. The expert-system development environment included in IMAGINE was used to develop a P index model, as well as the diagnosis and prescription expert systems (ES). Developed models were applied using the measured and observed data from the Vemmenhög watershed. A recommended Best Management Practice (BMP) for one of the fields identified as a "high potential for P movement" field was tested with the GLEAMS model.

Selection of soils

Preferential flow as a transport phenomenon occurs in both vertical and lateral directions. Its efficiency in vertical transport of different substances (such as nutrients and pesticides) has gained increased attention from a water quality conservation point of view. In a lysimeter experiment using Acid-red as a dye tracer, Bergström & Shirmohammadi (1999) showed the occurrence of a well-defined preferential flow pattern in undisturbed soil monoliths filled with a structured clay soil from the Lanna field. In contrast, flow in sand monoliths from the Mellby field was described in the same study as a more uniform, piston-flow-type solute movement. They also concluded that ponded flow conditions enhanced preferential flow and that the cross-sectional images indicated a lack of side-wall effects on solute movement. The soils used in the above-mentioned work, the Lanna clay soil and the Mellby sand, are two of the most well-described and documented soils in Sweden. They have previously been used for studies of pesticide (Bergström & Jarvis, 1993; Brücher & Bergström, 1997, *etc.*) and nitrate (Bergström & Johansson, 1991) transport. Using the MACRO model, Larsson (1999) tried to quantify the effects of macropore flow on pesticide and nitrate leaching from the Lanna clay soil. For all the above-mentioned reasons, Lanna clay and Mellby sand monoliths, and Lanna clay field plots were selected for the studies included in this thesis.

Results and discussion

Reading between the articles

The large number of processes in which P is involved, and the even larger number of factors affecting these processes require a more holistic approach when attempting to describe the P cycle in the environment. However, in order to estimate the significance of one or a few important factors, experiments often have to be conducted under controlled conditions where the influence of other factors is limited. The limitation with such an approach may be that processes are studied under short periods or experiments are carried out in small, controllable volumes of a given system (*e.g.* soil), such as laboratory columns, lysimeters or field plots.

To shift the scale to watershed level, DSS developed in a GIS environment was used as a systems analysis tool to account for spatial variability of important soil properties and management data information at such a large scale. This work also indicated that a different approach to the P issues was needed. The development of the DSS required a way of thinking much closer to that of farmers, agronomic advisers, and decision-makers, in which the focus was placed on problem-solving approaches and logical reasoning.

The importance of preferential flow pathways for P displacement from the topsoil to tile-drain depth was evident when comparing P loads from clay monoliths (average 4.0 kg ha^{-1}) with the P loads from sand monoliths (average 56 g ha^{-1}) (Paper I). The relatively large P sorption capacity in lower layers of the sandy soil in combination with lack of preferential flow pathways were the main reasons why added P did not leach through the soil profile. In contrast, in clay monoliths water and solutes were displaced through preferential flow pathways, bypassing the soil matrix with its high P sorption capacity. However, due to the "the worst-case scenario" conditions used in this experiment (high fertilizer application followed by intensive water application creating ponded conditions), the large loads observed from the clay soil should be considered as potential P losses under extreme climatic and environmental conditions and not as actual losses under field conditions. Lower P losses under natural climatic conditions (between 1.25 and 1.86 kg ha^{-1} during the period from July 1998 to May 2001, depending on management treatment) obtained in the study described in Paper II (Table 2) are more representative for field conditions. Phosphorus losses from lysimeters where P fertilizer was incorporated into the topsoil were significantly lower compared to P losses from lysimeters where P fertilizer was applied on the soil surface, regardless of whether conventional tillage or no-till was used (Paper II). In other words, destruction of macropores in the topsoil with conventional tillage was not sufficient to reduce P losses. In contrast, mixing and incorporation of P fertilizer into the topsoil horizon decreased P losses significantly. However, repeated high application of P fertilizer caused increased losses from the P incorporation treatment as well, emphasizing the importance of sustainability principles, where P additions should follow crop requirements. Even these P losses were somewhat higher then those observed in field-plot studies, such as the one described in Paper III. In that field study, the total P loads over the 4-yr period (1993-1997) varied between 0.421 and 0.805 kg P ha^{-1} (Table 2). However, P loads for these plots during the agrohydrological year 1998/1999 exceeded 1.0 kg ha^{-1} soluble reactive P following the application of 80 kg ha^{-1} of superphosphate in November 1998 (Ulén & Mattsson, 2001). The variation in P loads between years is, most likely, a reflection of the natural temporal variations in climatic conditions. The episodic pattern of P losses was a common denominator for all experiments. For instance, nine high-flow episodes from a field plot (Paper III) accounted for more than half of total P and sediment losses, but only for one fourth of the total discharge. High discharge episodes in connection with snow melting and soil-frost thawing were observed in the lysimeter study as well (Paper II).

Table 2. *Phosphorus losses from the Lanna clay soil investigated in 4 different studies*

	Type of study	P losses kg ha^{-1}	Observation period	Comments
Paper I	lysimeter	2.05-6.57	5 weeks	5 x 100 mm water applied under ponded conditions
Paper II	lysimeter	1.25-1.86	July 1998 to May 2001	Natural climatic conditions
Paper III	field plots	0.42-0.81	July 1993 to June 1997	Natural climatic conditions
Ulén & Mattsson (2001)	field plots	1.00-2.47	July 1998 to June 1999	Natural climatic conditions

The partition between different P forms in leachate from the lysimeters (Papers I and II) and in drainage from the field plots (Paper III) differed significantly. The main discrepancy was a higher contribution of particulate P to the total P losses in tile drainage from the field plots, whereas dissolved P represented more then 70% of total P losses in lysimeter leachate (Papers I and II). The dissolved P percentage in drainage was usually significantly lower and depended on the timing of P fertilizer application (Paper III). Under field conditions, erosion may be induced either by rain-drop effects or by water flow on the soil surface and, in the latter case macropores may act as "entry points" for the run-off water carrying detached soil particles and particulate P. This phenomenon is limited in lysimeters due to their small size, and instead of lateral run-off, ponded conditions are often created on the soil surface. Another possible explanation for a higher particulate P content in tile drainage may be the long-term accumulation of fine soil material near the drain-tiles (Ulén *et al.*, 1998), which is transported to the drains during flow events.

As mentioned earlier, Bergström & Shirmohammadi (1999) showed that ponded flow conditions enhance preferential flow as a physical process. Logan & McLean (1973) stated that constant head leaching resulted in greater movement of ^{32}P out of the surface layer and greater accumulation in the leachate than intermittent leaching. The observations in the lysimeter experiment described in Paper II showed that the highest P loads were associated with spring-flow discharge after rapid snowmelt periods and soil defrost. Snowmelt water often created ponded conditions on the soil surface prior to soil defrost. Jensen (2000) divided the subsurface P transport process into two steps: (i) the loading step, where net transfer of P occurs from the P source (solid phase) to the mobile solution, and (ii) the translocation step, where P is transported with mobile solution through the soil profile. Ponding not only enhances the translocation step by preferential flow transport, it may also extend the loading step by prolonging the contact between stagnant water and solute/sorbed P as a P source. Moreover, the anaerobic conditions created by ponding may reduce Fe(III) to Fe(II) causing a release of P to the soil solution. For instance, Young & Ross (2001) showed that the flooding caused significant release of phosphate to both porewater and floodwater, although the phosphate concentrations increase was much higher in the former. The ponding phenomena observed in the lysimeters may occur as well under field conditions. Lower parts of a field, and macro and micro depressions, especially in the presence of frozen soil horizons after the snowmelt period, are inclined to ponding. Also, ponded conditions may develop within the soil profile as perched "groundwater", i.*e.* above the plow pan due to the lower hydraulic conductivity of the plow pan layer. Therefore, the parts of a field (watershed) where ponded flow conditions may occur should be given proper attention when planning P management. The current practice of plowing in P fertilizer/manure is therefore questionable in cases when the soil profile has a pronounced plow-pan layer. By doing this, the P source is brought down to the location where preferential flow may be induced and enhanced by ponded conditions. Tillage operations, which are more efficient in incorporating P fertilizer rather than in displacing it to the plow-pan depth are preferable, especially knowing the positive effect of P fertilizer incorporation on P-loss reduction (Paper II).

Shifting the scale from undisturbed soil monoliths (Papers I and II) via field plots (Paper III) to watershed level (Paper IV) introduces more factors governing P behavior in the system. However, for P losses to occur, it is necessary to have the simultaneous presence of a P source and a P-transport pathway at all scales. It gives us an opportunity to identify the vulnerable parts of the watershed and focus our abatement efforts on them. DSS developed in this study classified only small parts (5-10%) of the total watershed area as high-risk areas regarding P movement. Site-specific probable causes and BMPs were also identified for each high-risk field. The reliability of the results obtained by DSS as a system analysis tool depends on the proper definition of the important factors for P behavior in the environment. Recognizing the importance of preferential flow for P displacement, and thereby P-index calculations and abatement strategies, should be the next step in DSS development work. Derby & Knighton (2001) showed that preferential flow occurs also at field scale, where snowmelt runoff infiltrated the soil profile via small topographic depressions after the frost thawed. The development of new techniques (GIS, remote sensing, artificial intelligence) and software systems (programs and models) will additionally help us to account for spatially and temporally variable factors, at all scales.

Limitations and interpretations of the results

Several drawbacks related to lysimeter studies in general, and preferential flow studies in particular are often quoted in the literature, some of which are listed and briefly commented on below:

- *Risk of side-wall flow*
  Both the tracer study with Acid-red (Bergström & Shirmohammadi, 1999) and the study with ^{36}Cl and H_3O^- (Bergström *et al.*, 1994) showed a lack of side-wall flow effects when using the same type of undisturbed lysimeters as those used in this work.

- *Artificial boundary conditions at the bottom of the lysimeters*
  A water-saturated zone at the bottom of a lysimeter and the interrupted capillary connection to the subsoil should be considered when comparing the results obtained in a lysimeter study with field results. However, the depth of the soil monoliths used in this work was *ca.* 1 m, which is a quite common drain-tile depth in Sweden. Therefore, the bottom boundary conditions of the lysimeters resembled field conditions quite well during drain flow periods. However during dry periods, the interrupted capillary connection to the subsoil may have caused more pronounced drying of the soil profile in the

lysimeters. The raised groundwater under field conditions may mix and dilute percolating water, which should also be considered.

- *The average hydraulic conductivity*
  Bouma (1990) stated that at least 30 structural units should be present in a column cross section to obtain a meaningful statistical average of hydraulic conductivity of a soil when using intact soil columns. It is questionable that the size of the lysimeters used here was large enough to capture 30 structural units, but studies showed that the size of the lysimeters was sufficient to capture the preferential flow pathways (Bergström & Shirmohammadi 1999), and their effect on P displacement (Paper I).

- *Blocked lateral surface and subsurface water flow*
  Lateral surface and subsurface water and solute flow cannot be properly simulated using lysimeters or even in field plots. Lysimeter and plot studies are useful in cases where the main flow is vertical. However, conditions in some parts of the field (such as depressions and lower parts of the field) may be more comparable to the conditions in the lysimeter.

These conditions show that a direct interpretation of the results obtained in lysimeter and field-plot experiments in terms of actual field conditions is not possible, especially not in quantitative terms. However, their importance for identification and evaluation of processes is essential.

The radioactive P isotope ^{33}P was very useful as a P source tracer (Paper I). The domination of added ^{33}P in leachate (on average more than 70%) observed in this study indicates a highly efficient P transport pathway connecting the soil surface, where the ^{33}P-labeled fertilizer was applied, and the bottom of the lysimeters (1 m), where leachate was collected. This high connective capacity of the soil surface and the depth of the drain tiles in a field are very important, considering that the bulk of added ^{33}P remained in the top 10 cm of the soil profile. The short half-life (25.4 days) did not create any problems because of the high intensity of transport processes driven by the large water applications. Additionally, the short duration of this experiment (four weeks) utilized the short half-life optimally. The episodic leaching pattern under natural climatic conditions (Paper II), with small leaching quantities during autumn, shortly after ^{33}P-labeled fertilizer applications, and high losses during the following winter-spring period, when the ^{33}P measurements were unreliable due to the low radioactivity values, exposed the shortcomings of the use of short half-life isotopes for long-term studies. Additionally, the gradual wetting of the soil surface during autumn, without triggering preferential flow, may also have promoted P sorption, and increased the influence of isotopic exchange processes (Di *et al.*, 1997).

The reliability of DSS developed in the study described in Paper IV depends on the input data and the set of rules created during the development of P index and

ESs. DSS should be considered as a dynamic system, where new knowledge must be incorporated within the system as ongoing research improves our understanding of the P issue. Without any doubt, there are a lot of possible adjustments and modifications that can be made to different parts of the DSS, but the approach of stepwise identification of critical source areas, and diagnosis of site-specific probable causes followed by site-specific BMP prescription and its model evaluation should be the guideline for further improvement in P loss abatement.

What have we learned?

The following points should be considered both as conclusions of this work and an overview of the ongoing work with suggestions for future efforts:

- Preferential flow is an important pathway for P transport through the soil profile. However, its efficiency in P transport depends on P-source availability and temporal/spatial variations in preferential flow occurrence. Therefore, the abatement should also be site-specific, targeting the most critical areas during critical periods. GIS could be used for identification of ponding-sensitive areas within a watershed/field. Sensitivity to ponding above surface and above the plow pan should be incorporated in P-index calculations.

- Added soluble fertilizer P is an important P source. Its incorporation in the soil rather than surface application enhances P sorption and reduces the negative impacts of the loading step of the P-transport process. Application of tillage practices that are more efficient in P incorporation and mixing into the soil can help us to utilize preferential flow to transport percolating water bypassing the soil matrix where the bulk of incorporated P fertilizer is stored. The current practice of plowing down surface applied P fertilizer should be closely examined due to the risk for ponded conditions above the plow pan. Store (basic) fertilizer applications with high fertilizer rates should be avoided as much as possible, especially in areas sensitive to leaching.

- The episodic character of P losses requires both site- and time-specific understanding of the factors governing increased P concentrations in leachate. In other words, we should avoid P applications (fertilizer/manure) at sites and during occasions when P transport (surface run-off or preferential flow) is likely to happen. Thus, the "P source-P transport" approach can be used in the abatement work as well, where the elimination of either one of these two factors may help us to prevent P losses. Sustainability criteria should always be considered, both from economical and environmental points of view, so that no more P is added than used by the crop.

- The relation between soil-P content and P losses in leachate is the focus of an ongoing experiment, in which four different P levels and five different soils from a long-term fertility experiment are being studied. The results should help us to define criteria for P-loss sensitivity determinations based on soil-P tests.

- The development of risk assessment tools and Decision Support Systems are valuable for an overview, processing and understanding of P problem-related issues, processes and factors of importance for P behavior in soil. Introduction of a P index better adjusted for Swedish climatic, hydrologic, and soil conditions should help, enhance and facilitate site-specific abatement efforts. The GIS technique helps us to better account for spatial variability. Hydrologic and nutrient models working in a GIS environment may further improve our understanding of temporal variations and processes of importance for P behavior in soil.

- Research at different scales is necessary for better understanding of the P-loss issue. However, the interpretation of results, especially in quantitative terms, should be carried out very carefully, recognizing not only the complexity of factors involved at different scales, but also the effects of the potential interplay of different factors. Therefore, an interdisciplinary approach is needed to develop a sustainable solution for the P issue.

Sammanfattning (Swedish summary)

Fosforförluster från jordbruksmark måste reduceras eftersom fosforn (P), som är det begränsande näringsämnet i sötvatten, kan intensifiera eutrofieringsprocesser i sjöar och vattendrag. Fokus beträffande forskning och motåtgärder har framför allt varit riktat mot ytavrinnings- and erosionsförluster av löst och bunden (partikulär) P. Höga P förluster i dräneringsvatten och förhöjda P koncentrationer i sjöar, vars avrinningsområden domineras av lerjordar, antyder att det även finns andra vägar för P transport. Markprofilens roll som en "buffertzon" och ett skydd mot P förluster har ifrågasatts i detta arbete. Markvatten som P bärare kan nämligen transporteras igenom en liten del av den totala porvolymen utan att komma i kontakt med huvuddelen av markprofilen. Strukturella makroporer i lerjordar, tillsammans med rot- och maskgångar, utgör genvägar för P förflyttning från matjorden till dräneringsrören. Två lysimeterförsök och observationer från fält utnyttjades för att klargöra makroporernas betydelse för P transport, samt för att testa olika motåtgärder. Slutligen utvecklades ett GIS-baserat beslutstödjande system som ett verktyg för identifiering av de känsligaste områdena inom ett avrinningsområde, samt för diagnostisering av de bakomliggande orsakerna till P förluster och rekommendation av den mest passande motåtgärden.

Resultaten visade att applicering av P handelsgödsel märkt med den radioaktiva P isotopen ^{33}P på markytan av en lerjord och en sandjord gav ett tydligt svar på makroporernas betydelse för P transport. P förlusterna från lerjorden, där makroporflöde var den dominerande transportmekanismen, nådde i genomsnitt 4,0 kg ha^{-1}, medan P förlusterna från den homogena sandjorden var i genomsnitt bara 0,056 kg ha^{-1}. Merparten av P utlakades i form av löst P. Förstörelse av makroporerna i matjorden genom "jordbearbetning" gav ingen signifikant minskning av P förlusterna jämfört med ingen bearbetning. Däremot reducerades P förlusterna signifikant genom inkorporering av P gödseln. En ökad kontakt mellan jordpartiklarna och gödseln hjälpte till att behålla P inom markprofilen. Ett episodiskt mönster av P förlusterna, där merparten av de totala P förlusterna sker under ett fåtal intensiva flödesepisoder, var den gemensamma nämnaren för både lysimeter- och fältförsöken. Förekomst av fritt vatten på markytan eller i markprofilen intensifierade utlakningsprocesserna genom makroporer.

Höga P förluster förekommer när både en P källa och en effektiv P transportmekanism är samtidigt närvarande, på en och samma plats. Denna princip kan utnyttjas för att identifiera de delar av ett avrinningsområde som är mest känsliga, och på så sätt fokusera motåtgärderna till dessa delar av avrinningsområdet där de har den största effekten. De fältspecifika orsaker som ligger bakom P förlusterna måste identifieras och motåtgärder måste grundas på lokalspecifika förhållanden. Istället för att se de temporala och rumsliga variationerna i P förluster som problem, måste vi försöka förstå de bakomliggande mekanismerna och grundat på detta göra en bedömning och urval av den mest effektiva motåtgärden. Nya tekniker som GIS och artificiell intelligens kan ytterligare förbättra bearbetning av rumsligt varierande data och möjliggöra en konsekvent och objektiv analys av de faktorer som påverkar P kretsloppet.

Sažetak (Bosnian summary)

Povećani dotok fosfora (P), kao hraniva koje limitira proizvodnju biomase u rijekama i jezerima, može izazvati njihovu intenziviranu eutrofikaciju. U posljednje vrijeme je pažnja sve više usmjerena na poljoprivredu kao jedan od najvažnijih eksternih izvora P, naročito nakon uvodjenja prakse prečišćavanja otpadnih voda. Do sada je naučno-istraživački rad bio usmjeren prvenstveno na probleme ispiranja P putem površinkog oticaja i erozije. Velike količine P izmjerene u vodi iz drenažnih cijevi, kao i povećane koncentracije P u jezerima u čijem okruženju dominiraju glinovita zemljišta ukazalo je na postojanje drugih, veoma efikasnih mehanizama za transport P sa poljoprivrednog zemljišta. Ova disertacija osporava ulogu zemljišnog profila kao zone koja svojim visokim kapacitetom za sorpciju smanjuje P gubitke, posebno u zemljištima u kojima se voda i u njoj rastvorena hraniva transportuju kroz tzv. makro pore u zemljištu. Na taj način se zaobilazi glavni dio zemljišnih čestica koje su aktivne u sorpciji P. Makro pore u zemljištu mogu biti biološkog porijekla (korijenje, zemljišna fauna) ali mogu nastati i kao posljedica širenja i skupljanja zemljišnih agregata, posebno u glinovitim zemljištima.

Dva experimenta provedena u lizimetrima, kao i mjerenja i opservacije u polju, su iskorištena za razjašnjavanje uloge makro pora u transportu P, kao i za testiranje metoda za redukciju gubitaka. U završnoj fazi je konstruiran kompjuterski model baziran na geografsko-informacijskom sistemu (GIS) i ekspert sistemima (vještačka inteligencija) koji bi trebao služiti kao sredstvo za identifikaciju najosjetljivijih dijelova jednog slivnog područja, kao i za davanje preporuka za najefikasnije protumjere. Površinska aplikacija superfosfata označenog sa radioaktivnim izotopom ^{33}P je nedvosmisleno pokazala mnogostruko veće gubitke P putem makro pora iz glinovitog zemljišta (u prosjeku 4 kg ha^{-1}) u poredjenju sa pjeskovitim zemljištem, gdje je nedostatak makropora omogućio sorpciju dodanog djubriva na zemljišne čestice. Inkorporacija i miješanje djubriva u površinski sloj zemljišta se pokazalo kao efikasna metoda za smanjenje gubitaka. Nasuprot tome, narušavanje kontinuiteta makro pora kroz površinsku obradu nije dalo željene rezultate. Ispiranje P kroz zemljišni profil je imalo epizodičan karakter, tj. najveći dio gubitaka se desio tokom malog broja intenzivnih epizoda. Uključivanje novih tehnika kao što su GIS i ekspert sistemi omogućuje nam bolji pregled i obradu informacija koje variraju u vremenu i prostoru. Identifikacija najosjetljivijih dijelova slivnog područja može povećati efikasnost poduzetih protumjera.

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