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Phosphorus in Tributaries to Lake Mälaren, Sweden: Analytical Fractions, Anthropogenic Contribution and Bioavailability

Riverine phosphorus (P) concentration and P-transport to Lake Mälaren, the third largest lake in Sweden, has been monitored for 35 years in 12 major tributaries. During a period of 15 months, complementary assessments of particulate P, suspended matter and dissolved reactive P were made. Particulate P comprised 64%, dissolved unreactive P 23% and dissolved reactive P 13% as flow-weighted means, with high seasonal variability. "Background" or "reference" P-transport in the streams were estimated by different methods and the anthropogenic contribution to P-transport was shown to be reduced over time. Potential algal availability of particulate P showed a mean availability of c. 45% for water draining arable and forested land, while the availability was higher for sewage discharge particulate P and algal P. Calculated total bioavailable P in tributaries was shown to equal reactive P measured on coarsely filtered water and it was indicated that the proportion of bioavailable P was higher during the period with high anthropogenic P-contribution than with a lower contribution.

INTRODUCTION

Phosphorus (P) plays a major role in the eutrophication of lakes, and the numerical coupling between P-input to lakes and eutrophication is well established (1–5). Therefore, monitoring of P-concentration and transport of phosphorus (P) in lake tributaries are fundamental to understanding and combatting eutrophication worldwide. In Sweden, regular monitoring has been carried out since 1965 in a major program addressing the eutrophication of L. Mälaren, the third largest lake in Sweden. Eutrophication problems and the research program were presented in the very first issue of *Ambio* (6), and are again covered in several articles in this issue (7–9).

The fractional P-composition analyzed in the monitoring program included tot-P (TP) and reactive P (RP) (Box 1), and the digestion and detection methods conform with current European standards (Box 1).

Three pieces of information on P input have been of special concern in the study of L. Mälaren.

- i*) concentrations and transports over time;
- ii*) concentrations and transports under "background" or "reference" conditions in order to reveal anthropogenic influence;
- iii*) bioavailability of ambient phosphorus.

During the formative years of the study, a number of papers by the late T. Ahl addressed the first 2 problems above (e.g. 10–12). In a later paper, Ahl (13) focused on the role of suspended particulate matter for P-transport under background conditions. Unfortunately, analyses of particulate P were missing in the regular program, and background concentrations could not be calculated as suggested by Ahl. In reviews at the time, (14, 15) and later (16), it was also pointed out that the latter 2 aspects were best covered with knowledge on suspended particulate matter and associated P.

However, in 1980–1981 this kind of information was gathered in a special study (17) which focused on P-forms in water

and where water from tributaries to L. Mälaren were used as examples together with a set of other types of water. This special study is presented here and includes: *i*) the basic composition of tributary P with comparisons to the results achieved by the routine method; *ii*) the role of suspended matter in P-transport, examined in some detail with complementary information on dissolved P; *iii*) information on bioavailable P from a special study within the L. Mälaren watershed (19) with calculation of potential bioavailability; *iv*) information from the special study used in a general discussion of methods to estimate "background" and anthropogenically derived P-transport in watercourses as well as the bioavailable P-fraction; and *v*) an indirect follow-up of background and anthropogenic fractions over time, and a comparison of estimates of the current situation with that from the special study.

LONG-TERM MONITORING OF P IN TRIBUTARIES OF L. MÄLAREN

The mouths of 14 tributaries entering L. Mälaren were chosen in 1965 for chemical monitoring to assess the impact of riverine input on the water quality of the lake (11). The strategy was to calculate the input of substances using monthly chemical samplings and daily to fortnightly assessments of water dis-

Box 1. Phosphorus variables and analytical procedures

Variables measured in the regular program (marked *) and those used in special studies of 12 inlets into Lake Mälaren in 1980–1981.

Variable	Filter	Treatment
1. Tot-P* (TP)	–	K ₂ S ₂ O ₈ digestion
2. Reactive P* (RP)	20 µm Munktell No 3 paper filter	Filtrate analysis
3. Total Dissolved P (TDP)	0.2 µm Gelman membrane	K ₂ S ₂ O ₈ digestion
4. Dissolved Reactive P (DRP)	0.2 µm Gelman membrane	Filtrate analysis
5. Particulate Phosphorus (PP)	0.2 µm Gelman membrane	Digestion on filter w. strong oxidizing acids
6. Suspended matter d.w.*	0.45 µm Sartorius 11 306	105°C, Electrobalance
7. Suspended matter d.w	0.2 µm Gelman membrane	105°C, Electrobalance

From the 7 analyses above, Dissolved Unreactive P was calculated as (DUP) = (TDP)–(DRP). Calculated Tot-P was obtained as a sum: TPsum = (PP) + (DRP).

In all analyses, reactive P was detected following the molybdenum blue method (18). During pretreatments (filtration, digestion) the digestion of regular samples was performed at 120°C (autoclave) with potassium peroxodisulphate (K₂S₂O₈) (20), whereas that on special samples employed a mix of perchloric- nitric- and sulfuric acid at 220°C (21).

Table 1. Watershed areas and land use of the studied 12 tributaries of L. Mälaren (22, 23).

	Total area (km ²)	Upstream lake area (%)	Arable land (%)	Forested land (%)
Eskilstunaån	4187	14.8	18.3	42.6
Arbogaån	3802	7	9.8	65.3
Hedströmmen	1058	8.1	7	71.3
Köpingsån	284	5.2	16	52.9
Kolbäcksån	3093	8.7	3.8	67
Svartån	754	3.9	18.6	57.2
Sagån	865	0.8	33.3	49.9
Örsundaån	727	1.3	31.3	48
Fyrisån	1982	2.3	22.2	55.7
Sävjaån	726	1.3	23	66
Märstaån	71	1	38	49
Oxundaån	271	7.5	18.5	31.9

charge. The water samplings were made at a depth of 0.5 m using a Ruttner sampler.

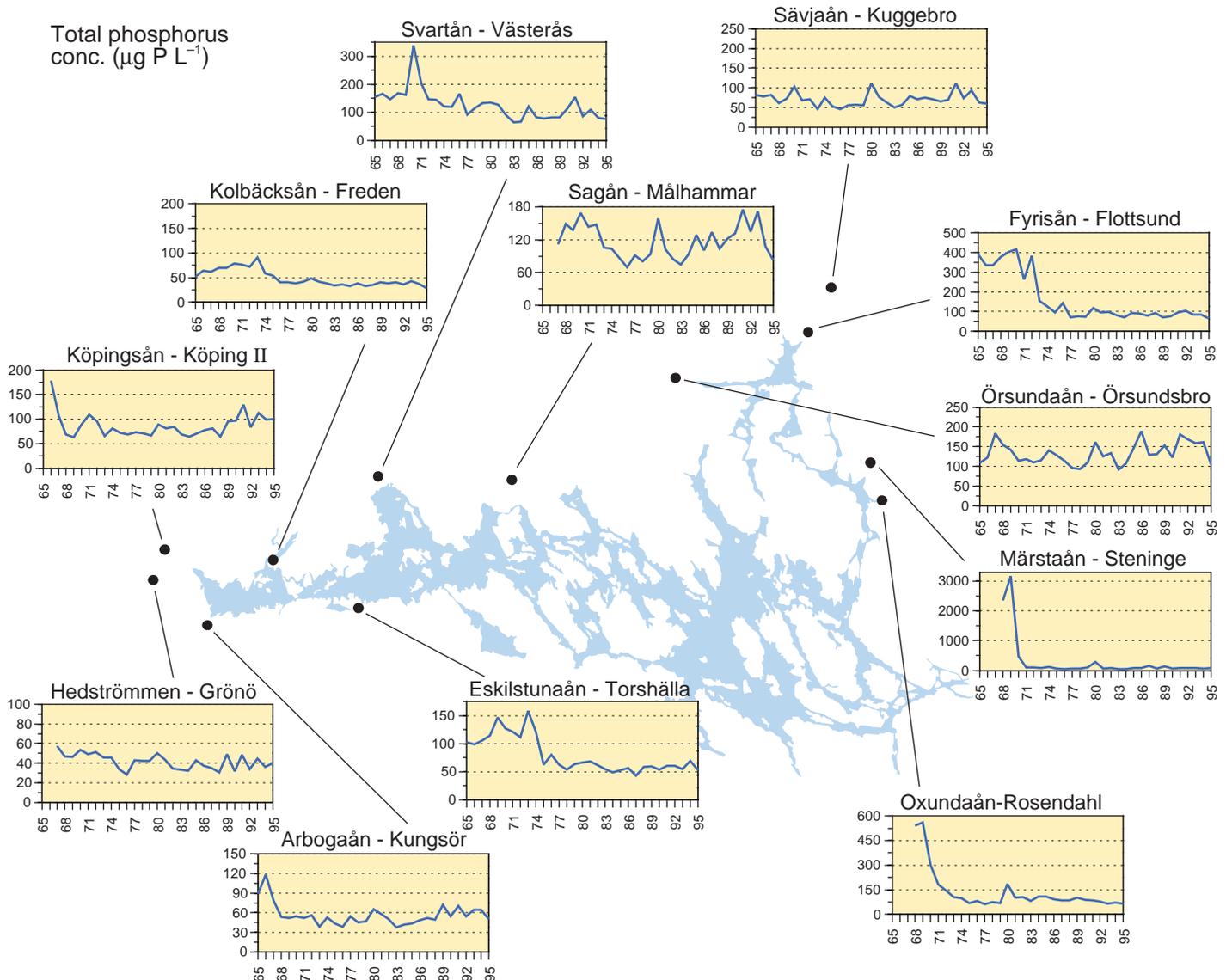
During the following 35 yrs the number of sampling stations were somewhat reduced, but monitoring has been continued at a core of 12 stations (Fig. 1) with essentially identical procedures carried out by the same laboratory until 1995, when sampling was regionalized.

The 12 inlets to L. Mälaren drain 79% of the lake's watershed (Table 1). The area-specific discharge varies from 6.5 to 10 m³ s⁻¹ km⁻² in a gradient from the lowlands around the eastern part of the lake to the higher terrain northwest of the lake. Upstream lakes are abundant in 7 of the tributary basins (4–

15% of watershed area) where the other 5 basins have less than 2% upstream lake area (Table 1). Together, the studied streams contribute 82% of the water input to the lake. The western tributaries to the lake are by far the most important for water input (cf. watershed area in Table 1), but input along the northern shore is also important for the receiving bays and basins.

During the initial 10 yrs, efforts were made to reduce the input of sewage phosphorus to the lake (22, 23). These measures

Figure 1. Annual mean concentrations of Tot-P at specified sampling stations in 12 tributaries of L. Mälaren. Generally, pollution abatement measures were put into action 1970–1975, in some cases earlier. Modified from Wallin (23).



were motivated among other things by the aforementioned impact models, primarily by that of Vollenweider (24); published 3 yrs after the tributary monitoring started. The municipal sources of P were cut by as much as 95% thanks to the introduction of chemical precipitation at the sewage-treatment plants. Phosphorus and nitrogen were also reduced in industrial outlets by forcing legislation. From the eastern shores of the lake 3 long tunnels were built to transport sewage, previously discharged into L. Mälaren, to the Baltic Sea c. 10 km to the east. One of the tunnels was not taken into operation until 1989, the others began operations in 1970 and 1974. Several of these measures were mirrored by falling inlet concentrations (Fig. 1). Other actions on point sources with outfall directly to the lake are not shown in the Figure.

After the initial clean-up, the conditions in the lake improved, and a new equilibrium between phosphorus input and lake status gradually evolved (8).

During the last 2 decades minor reductions in anthropogenic P-input have still been attained, but it is obvious that meteorological conditions are becoming increasingly important in regulating the P-input to the lake. It has also become evident that further measures of the P-input to the lake are needed to reach the even lower nutrient status desired by many concerned parties (25).

THE 1980–1981 STUDY OF PHOSPHORUS

Some 5 yrs after the major reductions in anthropogenic P-input, a study was initiated to determine which fractions of P were transported to the lake through different inlets. Some of these fractions (Box 1) can be traced to agricultural sources like soil loss due to erosion. Others are typically confined to waterborne humic matter, which is mainly derived from forests or mires. The sources of the other components cannot be identified. For some of these fractions, bioavailability can be approximated, which opens possibilities not only to assess the gross input of a fraction as described by chemical and physical properties, but also to indicate its biological importance for lake conditions. Thereby, a more efficient choice of future measures may be facilitated. Another purpose of the study was to compare the results of the fractionations to those of the routinely used monitoring fractions at the time.

Samples were taken over 15 months (March–May) in parallel to routine sampling at 12 of the stations used for monitoring (Fig. 1). The 15 months included 2 spring-flow events, and to make best use of data, means for the overlapping months (March, April, and May) were calculated before a yearly flow-weighted mean for each inlet was calculated.

MEAN FRACTIONAL COMPOSITION OF TRIBUTARY PHOSPHORUS

The levels of tot-P varied between 45 and 195 $\mu\text{g L}^{-1}$ as flow-weighted yearly means 1980–1981 at the sampling stations (Fig. 2). One stream, Märstaån, was extremely polluted during the first half of the study but recovered later. It is treated separately throughout this article. In the rest of the inlets, particle-carried P dominated (64%) the analyzed fractions, followed by dissolved unreactive phosphorus (23%), and dissolved reactive phosphorus (13%). Dissolved unreactive P (31% SD) showed the largest between-stream variation, followed by dissolved reactive P (22% SD) and particulate P (11% SD). The within-stream temporal variation was highest for particulate P (39%, mean of %SD within each station) followed by dissolved unreactive P (10%) and dissolved reactive P (8%).

The fractional composition can be compared to similar assessments within the Swedish network for assessment of nutrient losses from arable land which comprises 31 medium-sized drain-

age basins with typically > 50% arable land. Drainage from these basins had median portions of 50% particulate P, 14% dissolved unreactive P, and 42% dissolved reactive P in 1996–1998 (26). There is also a fairly high variability between the basins. Similar data from primarily arable land in the other Nordic countries provide similar information, both of variability and a fairly high portion of dissolved reactive P in stream water (27). The rivers

Figure 2. Concentrations of phosphorus fractions in water from 12 tributaries to L. Mälaren. Flow-weighted annual mean concentrations for the studied 1980–1981 period are given. The upper bars are derived from the special study, the middle bars from regular monitoring and the lower bars are derived from bioavailability data. PP = particulate P, DUP = dissolved unreactive P, DRP = dissolved reactive P, RP = reactive P (routine, with particles), TP-RP = difference routine total P-reactive P, PAP = potentially algal-available P.

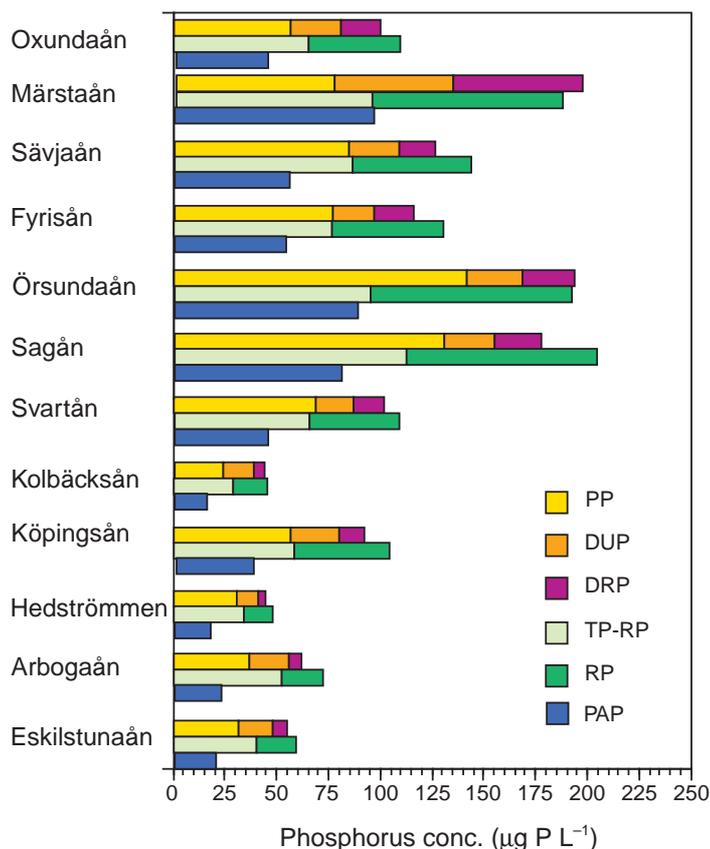
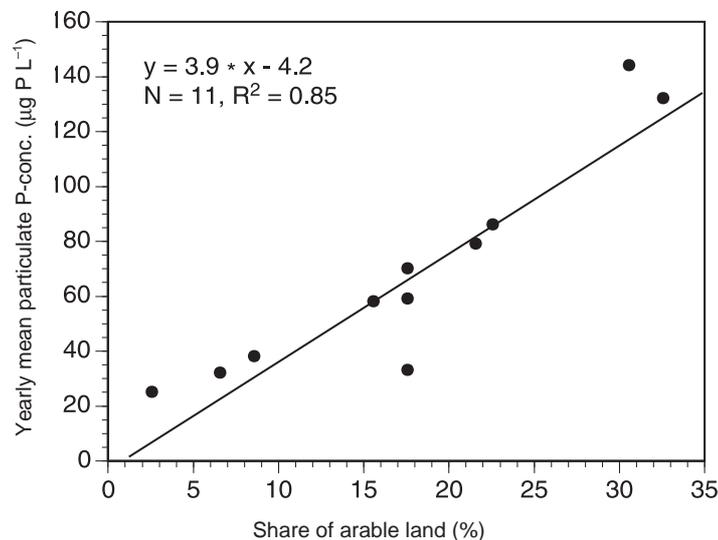


Figure 3. Flow-weighted annual mean concentration of particulate P for the studied period as related to the percentage of arable land in the L. Mälaren tributary watersheds.



flowing into L. Mälaren, on the other hand, seem to have both fairly similar fractional composition and a generally low fraction of DRP, possibly coupled to the lower share of arable land; a maximum of 33% only.

PARTICULATE P

Particulate P is highly important for the tot-P-concentration not only because it is the dominating fraction in the tributaries of L. Mälaren, but also because it is so variable over time. The linking to transport of suspended particulate matter is also obvious, which often is associated to water discharge, at least on a temporal scale. Therefore, the examination of particulate P occurrence in the watercourses starts with the coupling of particulate P to suspended matter and land use.

Both particulate P and suspended matter (using 0.2 µm filters) in L. Mälaren tributaries have a good fit to land-use variables in linear regressions (Märstaån excluded). The share of cropland area in the watersheds accounts for 85% of the variation in mean particulate P among the watersheds (Fig. 3) and explains 76% of the variation in suspended matter concentration. Among other land-use variables, only the percentage of upstream lake area contributes further to explain the variation in a multiple regression, while factors like the ratio between spring-sown crops to other crops (= inverted fraction of wintergreen fields) or livestock density are insignificant.

The equations describing particulate P and suspended matter concentrations as functions of the % arable land and upstream lake area are:

$$PP (\mu\text{g P L}^{-1}) = 32 + 2.9 \cdot \% \text{ arable land} - 3.5 \cdot \text{lake}\%$$

$$N = 11, r^2 = 0.93, p = 0.015$$

$$SS (\text{mg L}^{-1} \text{ d.w.}) = 16.2 + 1.9 \cdot \% \text{ arable land} - 2.8 \cdot \text{lake}\%$$

$$N = 11, r^2 = 0.85, p = 0.05.$$

Obviously, there is also a covariation between cropland-% and lake-% in the watersheds ($r^2 = 0.42$, $r^2 = 0.84$, with $N = 10$), but the pattern that emerges is consistent with the hypothesis that soil loss from cultivated areas is the main regulator of these concentrations. Concentrations are also to a large extent moderated by sedimentation in lake basins. Since particulate P is the dominant regulator of tot-P, there is also a good fit between tot-P and the above land-use variables. The share of arable land of the watersheds accounted for 87% of the variation in total P-concentration and if percentage upstream lake area is added, a total of 94% of the variation is explained.

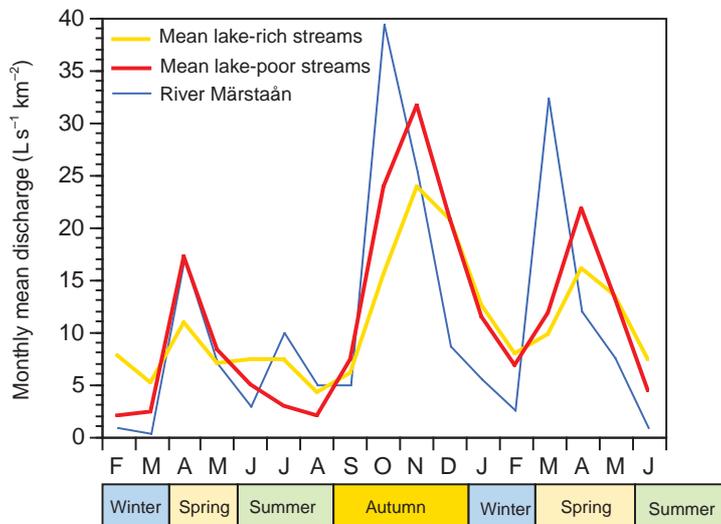
As to the sources of PP, inorganic and organic aggregates, mainly of soil origin, may transport P as part of their matrices, but also adsorb inorganic P from, e.g. pollution or fertilizers. However, losses from arable land also include pollution from the whole landscape including from countryside inhabitants and livestock. This contribution may be highly significant (27).

THE SUSPENDED MATTER AS A PHOSPHORUS CARRIER

Both content of P in suspended matter and suspended matter concentration in itself is coupled to temporal water-flow pattern and extent of flow in different years. Together they determine the particulate P present in the watercourses. In Figure 4a, the area-specific water discharge during the study period and adjacent months is presented. The division into 2 groups of streams (and the "outlier", Märstaån) is made so that streams with small water storage (< 2% lake area in the watersheds; Table 1) are separated from those with higher storage capacity and flow damping (4–14% lake area). The watercourses in lake-poor watersheds identified herein are identical to those with the highest cropland share (Table 1). These are Sagån, Örsundaån, Fyrisån and Sävjaån.

The hydrographs for the studied period suggest a division into seasons according to Figure 4. Support for this interpretation is

Figur 4a. Monthly water discharge Feb. 1980–June 1981 as means of streams draining lake-poor areas (the rivers Sagån, Örsundaån, Fyrisån and Sävjaån) and 7 streams draining lake-rich watersheds (the rivers Eskilstunaån, Arbogaån, Hedströmmen, Köpingsån, Kolbäckån, Svartån and Oxundaån).



Figur 4b. Monthly mean suspended matter concentrations in stream groups according to Figure 4a. Duplicate assessments on 0.2 µm membrane filters for the period April 1980–May 1981 are presented (bars).

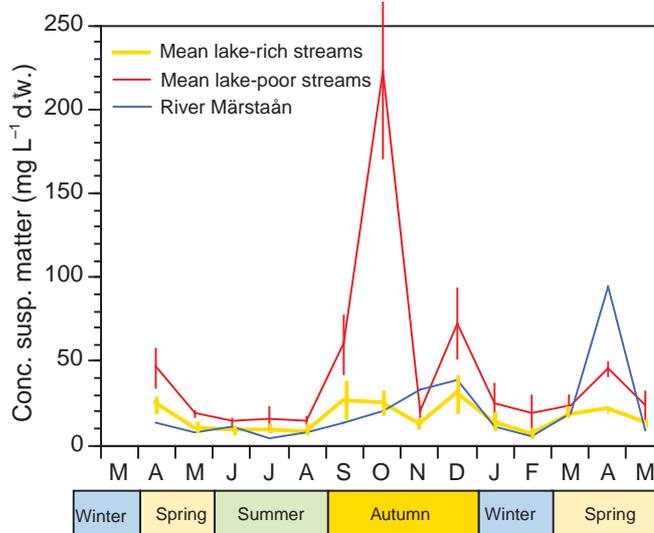
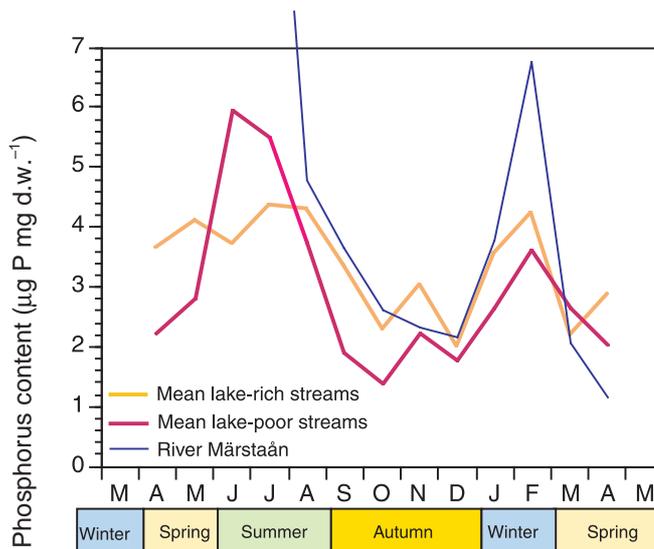


Figure 4c. Mean phosphorus content in suspended matter sampled on 0.2 µm membrane filters for stream groups according to Figure 4a.



also offered by the behavior of the studied variables during the period.

The 2 spring-flow pulses were fairly typical for the long-term monitoring period, whereas the autumn flow pulse was unusually high (Fig. 4a). Therefore, the yearly discharge was higher than normal. The discharge pattern typically consists of low flow during winter freeze, followed by an intensive spring-flow during spring thaw, low summer flow, and high and irregular flow during autumn rains, declining during winter freeze-up. Meteorological variations are large, however, and the timing and extent of flow pulses vary between years. In the monitoring period of L. Mälaren, winter freeze and low flow may even appear to be substituted by mild winters with high winter flow.

Flow damping in the lake-rich basins was particularly evident during periods of low discharge, but also during peak flow. The hydrograph for the smallest watershed, that of Märstaån (also lake-poor), shows a more dynamic pattern with a faster response to meteorological events.

Suspended matter concentrations (0.2 µm membrane filters) in rivers draining lake-poor areas exceeded that of the others by about one order of magnitude during the autumn peak flow and by about twofold during the rest of the period (Fig. 4b). Seasonally, suspended matter concentrations are known to be positively correlated to flow, or rather to yield a hysteresis curve vs water flow, i.e. the concentration rises at the front edge of a flow peak and declines more rapidly than the flow peak. Such patterns cannot be examined in detail here, but concentrations in the water of rivers draining areas poor in lakes and rich in croplands fit such a description (Fig. 4b).

QUALITY OF SUSPENDED MATTER

The P-contents of the suspended particulate matter span over one order of magnitude. The bulk of the samples (25–75 percentiles) have P-contents in the range 0.18–0.41% d.w. with a median of 0.27%. This is in accordance with a parallel study of particulate P (19) and also agrees with a number of other studies (review, (14)).

Märstaån, the outlier noted above, had extremely high P-concentrations in the suspended matter during the first spring-flow (mean 1.25%) and during summer (mean 1.8, peak 3.5%). The rest of the period (from August onwards) the P content was in the same range as for the other streams. As for the temporal variation of the phosphorus content of suspended matter (Fig. 4c) there is an indication of an inverse relation to suspended matter concentration. The seasonal variation of P-content however, is not as large as in suspended matter concentration. If the extreme P-content of the suspended matter of Märstaån is excluded, the P-concentrations generally vary by a factor 2 over the year.

For the quality of suspended matter, the general rule allows us to expect more heavy minerogenic material during high and turbulent flow. During base flow or low flow such material will usually be sedimented to the bottom of watercourses/lakes. Under such conditions, water mass particulates consist of finer and/or more organic material. Even live algae and bacteria may appear during summer.

Through a number of special samplings within the River Fyrisån drainage area (19), information was gathered on suspended particulates from specific sources like cropland ditches, forest brooks, the outfall from a chemical-precipitation sewage plant, and from somewhat larger watersheds with composite sources. The studies included centrifugations in 2 steps to separate easily sedimented suspended matter and more bouyant, yet filterable material. The centrifuge separation showed that the rapidly sedimenting fraction always had lower P-content and lower organic content than the slowly sedimenting fraction. The parent material, a marine calcareous clay, from the plain-land, had a P-content of 0.05–0.08% d.w. at 2.5% organic content (meas-

ured as loss on ignition), whereas the lowest P-concentration during autumn high flow in nearby Sävjaån was 0.12% at 3% organic content. This was also found to be the approximate baseline concentration at high flow in the streams draining cropland plains in the area. Higher P-concentrations in the analyzed riverine particulates were always coupled to higher amounts of organic matter. Such material seldom exceeded 40% organic content at the time 0.3 to 0.4% P was found in the material. Two important exceptions were noted; fresh planktonic algae had P-contents up to 0.9% and particulates from the sewage outfall had contents 3.5–4.3%, i.e. one order of magnitude higher than other riverine particulates. This is comparable to the common P-contents in household detergents at that time. It is thus reasonable to suspect that detergents were one of the sources of the particulate P in Märstaån. In fact, there was an upstream laundry service but no major municipal sewage sources (sewage was diverted in 1970). However, the exact source of the combined high particulate and dissolved P-losses have not yet been traced. Judging from the notes on P-content in planktonic algae one might suspect that the unusually high P-content of suspended matter during summer in the high-cropland streams is partly due to the appearance of planktonic algae at the river mouth sampling stations.

The analyzed P-contents of particulate matter are reported here as independent of the concentrations of P in solution. This holds true only for structural P built into organic matter, minerals or strong complexes. It is however quite likely that a part of the particulate P is in equilibrium with solution P, particularly dissolved inorganic P, due to sorption reactions (e.g. 28). It is also probable that particles in the watercourses have desorbed some P as compared to a pre-transport situation when they were surrounded by a more P-concentrated soil water as compared to the river water.

DISSOLVED PHOSPHORUS

A linear regression of flow-weighted means of dissolved P (TDP) on particulate P indicates that the 2 fractions reached equal size at low concentrations, but that total dissolved P only equalled one third of particulate P at high concentrations (Fig. 5). When means of dissolved reactive phosphorus (DRP) were regressed on PP, the picture was similar, indicating a DRP-concentration c. 20% of that of particulate phosphorus. The outlier, Märstaån (not shown), evidently had an excess of dissolved phosphorus not proportional to particulate phosphorus, as in the other rivers (cf. above). The abnormally high P-content in suspended matter from this stream indicates saturation of adsorption sites on particulate matter.

It should be noted in this context that the good coupling between particulate P and different forms of dissolved P seen in the present material is not at all the general rule, especially when it comes to streams draining watersheds with a high percentage of arable land and high concentration of total P. Among the 31 such watersheds cited above, total dissolved P was higher than one third of particulate P in all but one case, and the percentage dissolved reactive P of particulate P exceeded 20% in all but 4 cases (26). One might therefore suspect that higher amounts of P were sorbed to particles and that equilibria were approaching saturation or oversaturation for particulate P in many of these cases, whereas the amounts of sorbed P in L. Mälaren tributaries would be lower.

The calculated content of P in the particulate matter was also regressed on total dissolved P and dissolved reactive P for each sampling and station, in order to find indications that the P-content of the particulate matter might be regulated by some adsorption isotherm. However, no positive correlation between P-content of suspended matter and dissolved reactive P (or total dissolved P) was found. It appears that there was no common

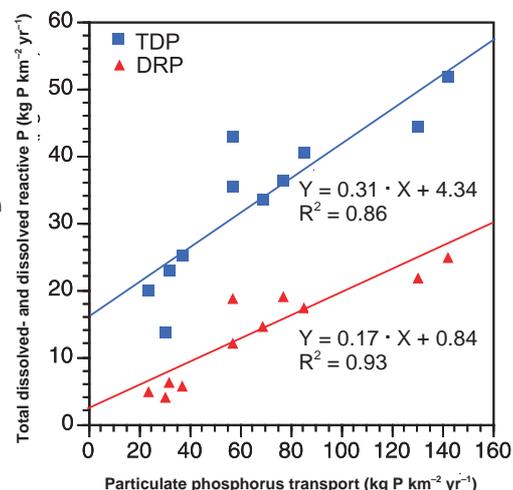
equilibrium concentration for all types of suspended matter, at least not during the specified conditions.

BIOAVAILABILITY OF TRIBUTARY PHOSPHORUS

The bioavailability of the transported P is an important issue which has been treated both in parallel with the present study (19) and otherwise over a long time period (review: 15, 16). Estimates of P-bioavailability, or rather P-availability for algae, are usually made through algal growth assays using the test sample as a P-source and calibrating against additions of pure orthophosphate. The term potentially available P (PAP) is often used for the outcome of these tests.

Such assays are often directed towards the bioavailability of particulate P, where the largest amounts of P are usually found, but at the same time a large variation in bioavailability is often seen.

Figure 5. Flow-weighted annual mean concentrations of dissolved reactive P (DRP) and total dissolved P (TDP) as functions of particulate P (PP) in 11 tributaries of L. Mälaren 1980–1981. The river Märstaån has been excluded.



Box 2. Comparison between special methods and routine methods

A. Suspended matter

The estimates of suspended matter mass discussed above, all refer to material retained on 0.2 µm membrane filters. These filters generally give higher estimates than filters of 0.45 µm porosity used in routine monitoring. A fixed conversion factor (0.45/0.2 µm) is however not recommended since the factor is constant at c. 0.92 (median, N = 46) only at high concentrations (in the interval 25–300 mg d.w. L⁻¹), whereas at lower concentrations the variation is large, around a median of 0.73 (N = 120). In the first instance, fairly large minerogenic particles, the smallest fraction of which is passing through 0.45 µm filters, are thought to dominate. In the second instance, a larger share of small minerogenic particles would be present and give a much lowered retention, specifically on 0.45 µm filters as compared to 0.2 µm filters. Also, organic aggregates may play a larger, but variable role, in this type of suspended matter. Finally, weighings in this low mass interval are known to be technically troublesome. This is at least reasonably well overcome by the duplicates used here for 0.20 µm filters.

B. Dissolved reactive phosphorus

When concentrations of dissolved reactive P (DRP) in membrane-filtered water (0.2 µm porosity) were compared to corresponding routine assessments of reactive P (RP) on coarsely filtered water (Munktell no. 20µ paper filter) the median of concentration ratios (RP/DRP) was 3.2 (Fig. 2). Expressed as a median percentage of tot-P, the 0.20 µm filtered DRP was 11% of the summation-tot-P (see below) and the paper-filtered reactive P was 33% of the routine TP. When inspected in a plot, the relation between the 2 types of RP assays was fairly variable and the regression equation has a high intercept:

$$\begin{aligned} \text{RP} &= 11.5 + 2.1 \cdot \text{DRP} \\ \text{N} &= 173 \text{ (6 obs } > 150 \mu\text{g P excluded)} \\ \text{R}^2 &= 0.51, \text{ p} < 0.0001. \end{aligned}$$

This raises the question of the source of the “extra” reactive P in the routine analyses. If it is derived from dissolved substances a maximum amount would correspond to the

analyzed TDP. This is unlikely to be the single source, however, since half of the values were found to be higher than the TDP and up to 3 times higher. It is most likely that P from particulate matter was analyzed by hydrolysis/desorption/extraction processes acting on the remaining particles passing through the coarse filter paper (cf. 29, 30).

The possibility that there is an analytical overestimation cannot be overruled. However, similar results to these have been obtained in a Norwegian study of riverine phosphorus in which reactive P of unfiltered water was compared to that of 0.45 µm filtered water and to that of GF/C glass-fiber filtered water (30). The Norwegian study found that the difference between unfiltered and filtered reactive P was large and significant.

C. Tot-P as a sum of particulate and dissolved-P

The procedures used in the analyses of particulate and dissolved phosphorus in the special study justify the summation as a means to acquire tot-P. As a matter of fact, the digestion of particulate matter with oxidizing acids is expected to give a somewhat higher P recovery than the persulfate digestion used in the routine analyses of tot-P.

When results were compared, the median of calculated quotients (routine TP/summed TP) was 1.06. Compared at a watershed level the routine mean tot-P was higher than summed TP in 8 streams, equal in 3 cases and lower in 1, (Fig. 2). A closer examination of a plot of the different assessments indicates that there is an intercept, “a blank”, for the routine measurements of c. 12 µg P L⁻¹, whereas the slope is 0.93 for the whole data set. In spite of the intercept, the expected lower recovery by the routine method may therefore still be valid. The suggested conversion function is similar to that of DRP:

$$\begin{aligned} \text{TP}_{\text{regular}} &= 11.5 + 0.93 \cdot \text{TP}_{\text{sum}} \\ \text{N} &= 163, \text{ R}^2 = 0.92, \text{ p} < 0.0001 \end{aligned}$$

Bioavailable dissolved P sometimes falls short of, but often equals, the fraction analytically determined as dissolved reactive P (15, 16, 28, 31–33). There is, however, general agreement on the low algal availability of P from the dissolved unreactive P-fraction. In most cases, the bioavailability of the dissolved unreactive P-fraction is close to zero (15, 16, 28, 31–34) with possible exceptions for dissolved unreactive P in sewage effluent water, which may support algal growth (16, 35). Consequently, 100% bioavailability for the dissolved reactive P-fraction is used here as the best estimate of bioavailable dissolved P, whereas 0% bioavailability is assumed for the dissolved unreactive P-fraction.

In Persson's (19) study, bioavailability of particulate P from Sävjaån and Fyrisån, draining the eastern part of the L. Mälaren watershed, was measured on 6 occasions and bioavailability of particulate P from subwatersheds draining cropland and forested areas within the same watersheds was studied. The bioavailabilities (PAP) for the 3 groups were 41%, 41%, and 55% for particulate P of riverine, cropland, and forest origin, respectively. Availability of lake seston P and particulate P in sewage effluent was even higher, i.e. c. 50–100%. All assay data have been pooled here in order to enable examination of the relation to P-content of particulate matter (Fig. 6). Tests with two different algal species (*Chlamydomonas* sp. and *Aphanizomenon flos aque*

uae) are included as well as tests on a rapidly sedimenting fraction and a more boyant fraction of suspended matter, separated by stepwise centrifugation (19). This type of presentation underlines primarily that suspended matter with higher P-content introduces more bioavailable P, whether the availability is 35 or 40% or whether it comes from forest or arable land (pooled in the Figure). Because there were no significant differences between the bioavailabilities for sample origin or for test algae, a mean of 45% algal availability was used for all particulate P in tributaries to L. Mälaren (except Märstaån; see below).

This availability was at the high end of those previously reported, where availabilities of 20–30% were frequent (see refs cited above). On the other hand 40–50% and 17–66%, availability was reported for tributary water (36, 37). A mean availability of 40% was also reported from Norwegian cereal cultivation fields (38). While there may be slightly conflicting bioavailability records for lowland streams, there is at the same time a consensus that particle-associated P from municipal sewage outlets (chemically treated or untreated) always have higher bioavailability than P from ordinary stream suspended matter, often by as much as a factor 2 (16, 19, 35, 39). Therefore, a bioavailability of 100% for Märstaån particulate P has been used herein. This is the only discernible case where phosphorus was carried by this special type of suspended matter. One might also

Figure 6. Content of potentially bioavailable phosphorus in suspended matter (y-axis) plotted on content of phosphorus in suspended matter. Particulate matter was sampled by 2-step centrifugation and tested in algal assays according to Persson (19). Data from different types of running water and 2 different test algae (*Chlamydomonas* sp. = quadrats and *Aphanizomenon flos aque* = triangles) are pooled. Lines indicate different relative bioavailability.

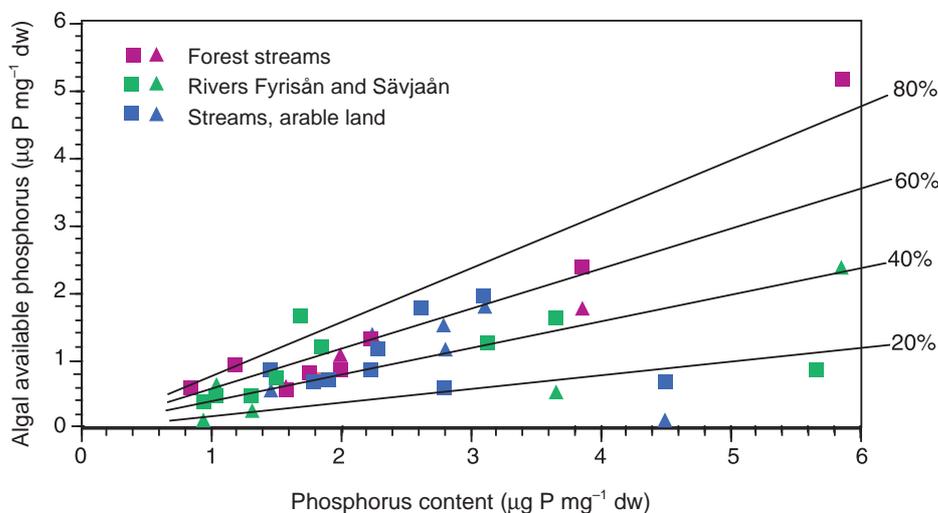
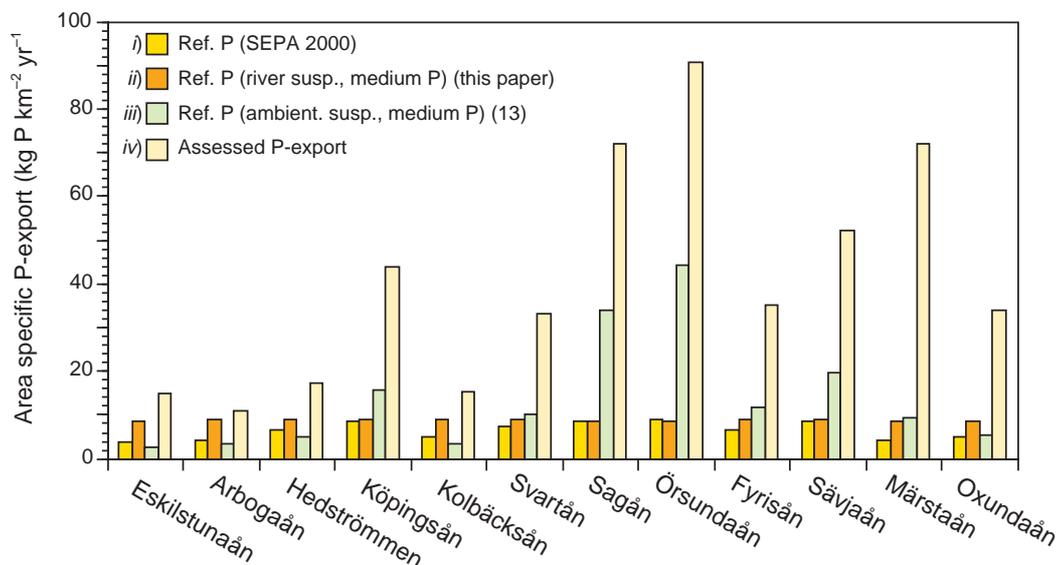


Figure 7. Different types of calculated “background” or “reference” values (i–iii) as compared to assessed yearly area-specific loss of total phosphorus from L. Mälaren tributaries 1980–1981 (iv). The Environmental Protection Agency's method (i) was compared to Ahl's method in which current transport of suspended matter is maintained, but phosphorus content of the particles kept medium high (iii). A third alternative presented here includes low and constant loss of particulate matter as in forest rivers with differential phosphorus content of particles from forest- and arable land (ii).



assume, that part of the particulates of Fyrisån is of this type, and that 45% bioavailability for particulate P in this river might be a conservative estimate.

In some reports (29, 30, 33) it has also been argued that the reactive P assayed on unfiltered water samples may provide a reasonable approximation of bioavailable P, whereas others did not find the same correlation (e.g. 16). For L. Mälaren tributaries there is an almost 1:1 relationship between the concentration of bioavailable P calculated along these lines and the directly reactive P (RP) analyzed with the present modifications (Box 1). The function was:

$$\text{PAP} = 0.91 \cdot \text{RP} + 3.1; R^2 = 0.97, N = 12, P < 0.0001.$$

This is a good fit, but we have to consider that estimates of bioavailable P in tributaries to L. Mälaren are few, and upscaled by using the same general constants throughout. Therefore, the fit is not definite proof, but a good indication. However, this treatment opens possibilities to predict bioavailable P both earlier and later temporally in L. Mälaren tributaries, since the RP analysis has been run with minor modifications throughout the 35 yrs (cf below).

BACKGROUND CONCENTRATIONS AND ANTHROPOGENIC INFLUENCE

Methods of estimating background P-concentrations or area-specific background P-losses are given in *Swedish Water Quality Criteria* (40), recommending that estimations of “reference” or “standardized background” P-losses are made using 5 methods based on:

- i) upstream lake percentage of the watershed;
- ii) area-specific discharge;
- iii) area-specific loss of COD_{Mn} ;
- iv) area-specific loss of silica;
- v) area-specific loss of humic substances (assayed as light absorbancy in water).

These functions each give low estimates for “reference” values and it is prescribed to use the highest value obtained for each stream. It is also noted that the first 2 approaches give fixed reference data with no temporal variation whereas the latter 3 give time dependent or dynamic values, basically ruled by meteorological conditions.

Since all data needed were available for the L. Mälaren tributaries (for 1 yr instead of the recommended 3 yrs) they were applied in calculations of reference P-losses for the “time weighted” 1980–1981 data. The results (Fig. 7) gave “background” P-losses of 3.4–8.8 kg P km⁻² yr⁻¹ for the 12 watersheds. In all but one case, they were derived from the COD_{Mn} -function.

There are other possible approaches. Ahl (13), in his paper on *Background Yield of Phosphorus from Drainage Area...*, discussed the role of suspended matter transport for P-transport and suggested a simple rule of thumb to obtain background P-yield by “multiplying the suspended sediment transport by the factor 0.001” (i.e. the P-content of the suspended matter was set to 0.1% d.w.). Estimates acquired by applying Ahl’s “rule of thumb” gave highly variable figures (2.6–44 kg P km⁻² yr⁻¹ for the streams due to the variable suspended matter transport. It should also be noted that Ahl did not include or mention dissolved P in his crude estimate, maybe he simply compensated by a somewhat high P-content of the transported material.

Data on suspended matter, particulate P and total dissolved P obtained in this study (Box 3) could be used as alternative ways of calculating the “background”. Erosion was mainly considered to be of anthropogenic origin and suspended matter exports from 15 Swedish forested watersheds (41) were used for calculations of reference export of suspended matter. Differential P-contents in suspended matter exported from forested areas and virgin

Box 3. An alternative “reference” or “background” approach for P-transport

According to this alternative approach, erosion in these plain areas is basically introduced by human activities. Thus, a reference value should be based on a strongly reduced suspended-matter transport from arable land; reduced to levels seen in forested watersheds. Nilsson (41) found a median loss of 1.5 tonnes km⁻² yr⁻¹ d.w. and a median organic content of 35% of d. w. in 15 major Swedish rivers mainly draining forested watersheds. Since P-content of the suspended matter was found to be proportional to the percentage of organic matter (19), 35% organic matter would correspond to a P-content of the suspended matter of c. 0.2%. A loss of 1.5 tonnes d.w. would then correspond to 3.0 kg PP km⁻² yr⁻¹. A somewhat rough—and maybe high—estimate of the corresponding total dissolved P for a given particulate P-concentration was obtained from the relation between particulate P and total dissolved P (Fig. 5), recalculated for transport data. The losses of tot-P after addition of TDP would then be 9.4 kg P km⁻² yr⁻¹. This area-specific P-loss has been ascribed to the forested parts of the watersheds.

For the cropland areas surrounding L. Mälaren, the content of organic matter of the cultivated soil layer is low, 3–5%, as compared to 20–30% for the upper layers of forest soil (42, 43). Such low contents of organic matter are rarely seen in particulates leaving cropland areas in streams, a fact which is probably due to a selective erosion of lighter and more organic particles, even during peak flow. A reasonable approximation of the composition of this material under “background” conditions would be c. 10% organic matter and a P-content of c. 0.1% (the same percentage used by Ahl). Thus, the PP-losses from this hypothetical virgin plainland would be lower than losses from forested areas and 1.5 kg PP km⁻² yr⁻¹ at equal transport of suspended matter. This would give a total P-export of 7.5 kg P km⁻² yr⁻¹ from this type of land with the maximum possible additional total dissolved P added according to Figure 5 (recalculated for transport data).

plains were set to 0.2% and 0.1%, respectively, guided by the organic content of the material. The export of particulate P was calculated and finally the total dissolved P corresponding to the given particulate P-concentration was obtained from the relation between particulate P and total dissolved P (Fig. 5) recalculated for transport data (Box 3).

Thus, particulate P-losses from the hypothetical virgin plains would be lower than losses from forested areas (1.5 as compared to 3.0 kg PP km⁻² yr⁻¹) at equal transport of suspended matter. This may appear controversial, but support for this reasoning was also found in the 31 studied Swedish cropland watersheds (26) where 3 of them had P-contents below 0.1%, the 25% percentile was at 0.16%, and the median was 0.25%. These losses represent losses from today’s cultivated and fertilized soils, and P-contents must be lower under background conditions.

Calculated for the different shares of cropland and forested land of the L. Mälaren tributary watersheds (Table 1) this reference approach would give losses of 8.5–9.0 kg P km⁻² yr⁻¹. By this reasoning there are now 2 alternative estimates of back-

ground or reference P-transport available in addition to those calculated following the SEPA-manual (40) (Fig. 7).

ANTHROPOGENIC INFLUENCE

When all different background estimates are compared for the different tributaries of L. Mälaren (Fig. 7) it is obvious that most approaches converge in the case of drainage from forested land (Table 1). The new approach presented in this paper gives higher estimates than the other approaches in tributaries with highly forested drainage basins, which was foreseen due to the fairly high P-content assumed for the particulate matter from virgin forest areas and maybe high TDP estimates. On the other hand, the construction of SEPA's standardized background calculation deliberately aims at alternative low estimates, of which the highest is to be chosen. The construction of a new estimate therefore, is not prone to give falsely high estimates and certainly will promote discussion.

When it comes to background estimates for cropland-rich areas, calculated reference P-losses diverge as expected. When Ahl's "rule of the thumb" including ambient concentrations of particulate matter (estimate *iii*; Fig. 7) is compared to actual P-losses, the difference demonstrates the effect of high concentrations of P in ambient suspended matter. From Figure 7 it is evident that elevated P-content in suspended matter contributes in particular to the high P-transport in Märstaån and Oxundaån. When discussing remedial measures, this is one important element of information.

Since no data on suspended matter are used in the derivation of the reference values according to the national criteria (40), the difference between the national reference values and the set of background data based on current suspended matter concentration (estimate *iii*; Fig. 7) points to the important role of the suspended solids *per se* for the P-losses from the watersheds. This difference between different background estimates points to the important role of suspended matter, in particular in Örsundaån, Sagån, Sävjaån and Köpingsån in decreasing order. This is another important element of information brought about by using many different background estimates.

The anthropogenic component of the P-loss can easily be estimated by subtracting reference values from the assessed values (Fig. 7). The degree of anthropogenic perturbation may also be visualized by computing the ratio between current loss and background or reference loss. Both may give guidance on the following issues:

- i) in which watershed effects of remedial measures may be expected;
- ii) point out watersheds with particular needs and/or possibilities for reductions of losses;
- iii) indicate what is to be gained from reduced erosion.

APPLICATION TO THE EARLY 1990s

The 5-yr period between 1991 and 1995 has been used to represent current status of the P-transport in L. Mälaren tributaries to which the 1980–1981 status was compared. Water discharge during both periods was higher than the 35-yr mean. During the early period, the annual mean was higher by 22% and during the late period by 17%.

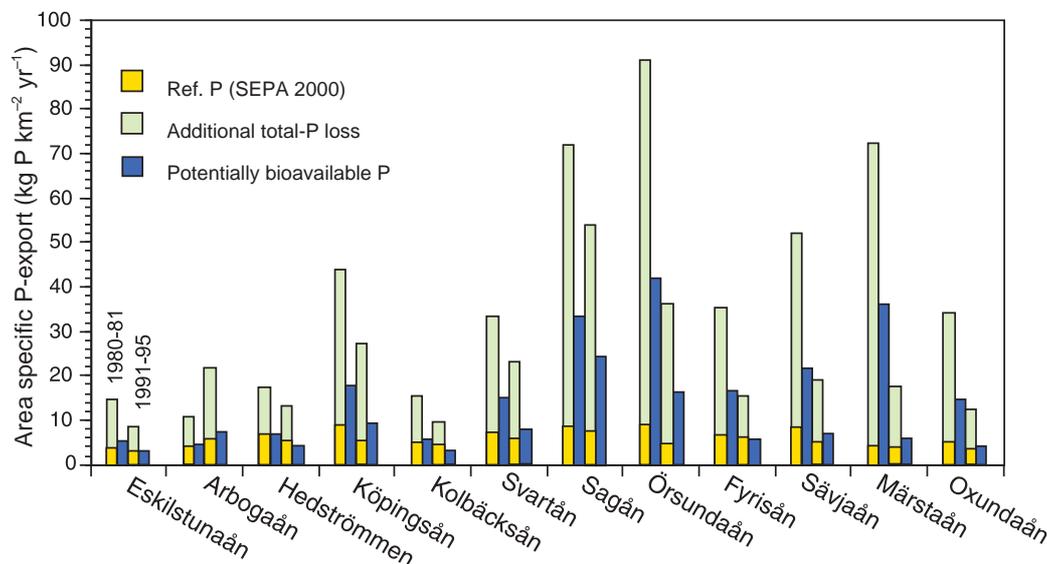
Calculated P-exports from the watersheds in 1991–1995 were considerably lower than those in 1980–1981 (Fig. 8). The mean ratio between late and early P-losses was 0.65, but in one case (Arbogaån) an increase was seen.

A part of the general decrease is caused by meteorological variations. By choosing the time-dependent dynamic type of background calculation according to SEPA (40)—with COD_{Mn} -transport as the governing factor—the effect of meteorological differences on reference values was experienced. The mean ratio in reference transport between 1991–1995 and 1980–1981 was 0.80, which explains why a significant part (c. 50%) of the transport difference between the periods was of meteorological origin. The increase in Arbogaån was also detected in reference values and can therefore be explained by natural changes.

An interesting question is to what extent the transport of bioavailable P has changed. Since this fraction was shown to be of almost exactly the same size as the reactive P-pool of the samples in 1980–1981, the transport of reactive P during the 1991–1995 period was used as an estimator of bioavailable P. The mean ratio between the bioavailable P for the late and early periods was 0.54, which indicates that this component had changed more than both tot-P and reference P.

A final question about the bioavailability of P-transported under reference conditions may find its answer in the studied material. Since particulate P is treated here as the main regulator of tot-P, and there is a general positive coupling to both total dissolved P and dissolved reactive P (Fig. 5), very low concentrations of particulate P would mean a dominance of dissolved unreactive P, which has been considered here as biologically unavailable. Thus, it appears that the original P-transport in the tributaries also had lower bioavailability and that the anthropo-

Figure 8. Comparison of area-specific losses of total phosphorus from watersheds (assessed), "reference" P (calculated according to SEPA (40) and potentially bioavailable P (see text) for the time periods 1980–1981 and 1991–1995. For each watercourse the first 2 bars show data for the former period while the last 2 bars show data for the 1991–1995 period.



genic contribution not only supplied more tot-P to the watercourses, but also supplied P of a higher bioavailability than the original P-input.

CONCLUSIONS

The 1980–1981 special study resulted in both a better understanding of the fractional P-composition in the tributaries and an opening for calculations of 2 types of “background” P-transport based on the transport of suspended matter. The approach that relied on assessed transport of suspended matter which was given a fixed P-content (13) was informative as regarding the role of suspended matter for P-losses. On the other hand, it was unrealistic since the currently assessed suspended matter certainly was anthropogenically derived to a large extent. The other approach suggested relied instead on assumed constant and low amounts of transported suspended matter with fixed P-content dependent upon the sources of particulate P. This approach has the possible advantage of not needing any data monitoring. On the other hand a better tuning of the constants to the conditions of specific watersheds may be needed. This is a disadvantage, but the point made here is merely to promote the use of differ-

ent estimates of reference or background transports of P, thereby giving an insight not only into the magnitude of the P-excess, but also into the causes and possible remedies for the exceedingly high P-transport.

The versatility of using a reference or background estimate that is variable over time was shown when the anthropogenic impact during different time periods was estimated. It is to be noted that 3 of the 5 alternative approaches recommended by the SEPA (40) are of this type, a fact which certainly promotes their use.

The actual reduction of P-input into L. Mälaren over time should be noted, especially during the first 10 yrs of the monitoring period (Fig. 1), as well as the reduction of the bioavailable fraction. According to most observations and general models (1, 3–5), this would have reduced the P-level and eutrophication of the lake, which has indeed been proved by monitoring and modelling (8, 22, 23) and indicated by biota (9).

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