PARTICULATE AND DISSOLVED LOSSES OF NITROGEN AND PHOSPHORUS FROM FOREST AND AGRICULTURAL SOILS

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Particulate and dissolved losses of nitrogen and phosphorus from forest and agricultural soils

by Richard A. Marston

Nitrogen and phosphorus generally exert the greatest impact on water quality among the various macronutrients, and an enormous literature exists on the intracycle transfers and overall budgets for these two nutrients. However, there are few studies of the relative amounts lost in dissolved and particulate forms. The transfer of nitrogen and phosphorus in these two forms represents a key link between terrestrial and aquatic ecosystems that governs the trophic state of receiving waters. The magnitude and timing of nitrogen and phosphorus export also responds to land-management activities (Likens and Bormann, 1974). The loss of nitrogen and phosphorus in either dissolved or particulate form amounts to a reduction in soil fertility. The resultant decline in vegetation cover creates a positive feedback in the ecosystem by facilitating greater erosion and loss of nutrients in the particulate form (Anderson, 1988). Comparisons between studies of nutrient export are made difficult by confused nomenclature and contrasting methods of reporting data. Some studies reporting on losses of nutrients examine only surface runoff and ignore the export via subsurface waters. Most studies which distinguish between dissolved export and the fraction associated with sediment have drawn on stream data from agricultural catchments. However, the application of results from agricultural to forest catchments is limited by the difference in dominant hydrologic processes and land-management practices peculiar to each cover type, as well as the fact that silvicultural activities are known to have differential effects on the release of sediment as opposed to nutrients (Stevenson, 1986).

Nitrogen and phosphorus losses from soils occur as both mineral particulate and organic particulate matter. Nitrogen and phosphorus ions may be adsorbed to inorganic particulates, but they may also become part of the composition of organic particles. The principal forms of nitrogen considered in studies of nutrient export by hydrologic pathways are gaseous ammonia (NH_3), the ammonium ion (NH_4^+), and nitrate (NO_3^-). The conversion of nitrite (NO_2^-) to nitrate occurs at a faster rate than conversion of ammonium to nitrite, resulting in low amounts of nitrite in soil. Consequently, NO_2^-, is not usually considered in export studies. NH_4^+ and NO_3^-, however, can be associated with sediment or found in dissolved form. With regard to phosphorus, orthophosphate (PO_4^{3-}) is most important,

occurring in organic or inorganic form as $H_2PO_4^-$ or HPO_4^{2-}. The division of dissolved and particulate nutrients is usually based on separation by a filter, with inconsistent pore sizes reported between 0.42 and 0.45 micrometers (Ryden *et al.*, 1973). Further, the dissolved and particulate components of nitrogen and phosphorus can each be divided into inorganic and organic factions (Figure 1).

A schematic diagram of nutrient pathways to streams helps to illustrate the sources of particulate and dissolved nutrient releases (Figure 2). Organic particulates and dissolved organic compounds are generated from the nitrogen and phosphorus content in organic matter in the plant and organic biomass residues. The dissolved inorganic component is released from the available nitrogen and phosphorus, that is, the portion of those nutrients which can be absorbed by plant roots (i.e., the dissolved component). The inorganic particulate component involves minerals derived from geochemical weathering of parent material, principally apatite in the case of phosphorus.

The following discussion of particulate and dissolved export of nitrogen and phosphorus is organized around four major topics. First, a brief review of intrasystem cycling of nitrogen and phosphorus cycles is presented, with emphasis on the processes controlling the relationship of nitrogen and phosphorus with sediment. Secondly, the reported sediment-nutrient relationships established for nitrogen and phosphorus export from agricultural catchments are discussed. Thirdly, introgen and phosphorus export studies on forested catchments are reviewed which distinguish between the dissolved component and particulate

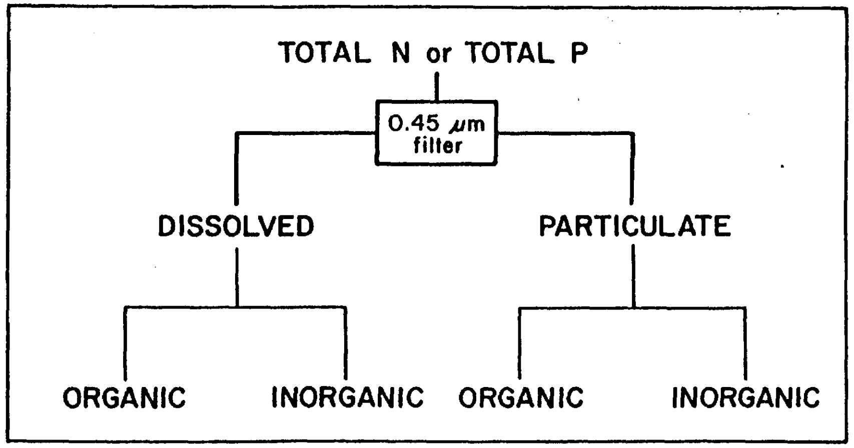

Figure 1 The distinction between dissolved and particulate nutrients in the present study.

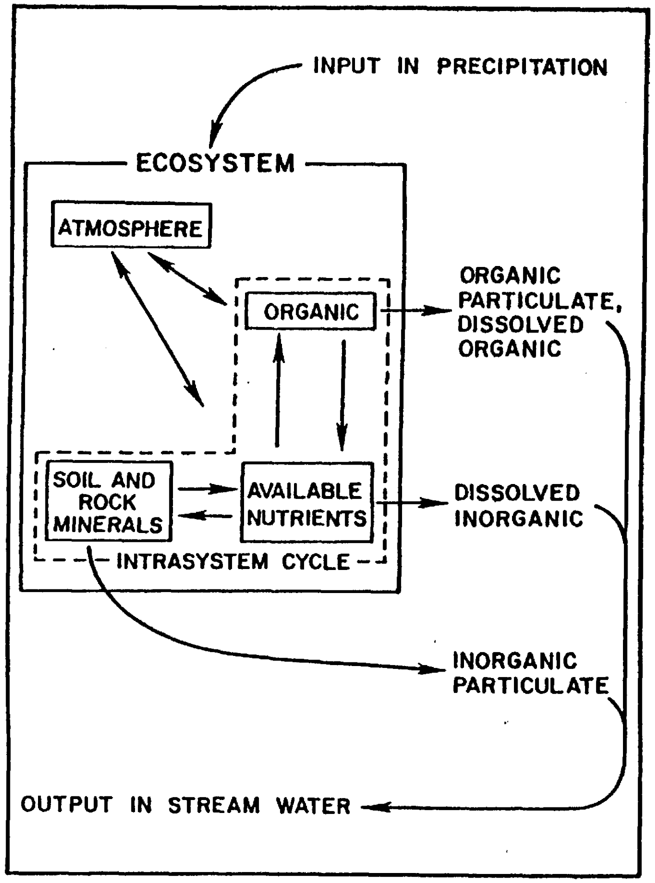

Figure 2 Pathways for particulate and dissolved forms of nitrogen and phosphorus (after Bormann, 1969).

component, with a concurrent discussion of the impact of silvicultural activities in changing the relative importance of the dissolved and particulate nutrient transport processes. Finally, two nonpoint source models sponsored by the US Environmental Protection Agency (EPA) are criticised with respect to their value in using sediment-nutrient relationships to predict nitrogen and phosphorus

export from forested catchments. These two models deserve particular attention because of their widespread application. Other models are reviewed by Novotny and Chesters (1981). A detailed account of other forms of nutrient loss from soils, including bacterial-chemical denitrification and volatilization, are beyond the scope of the present review, but again, readers may refer to Novotny and Chesters for a review.

I Intrasystem cycling of nitrogen and phosphorus

The relative importance of particulate transport of nitrogen and phosphorus as opposed to transport by solution depends on the intrasystem cycling process for each of these nutrients, and their adsorptive properties in soils and stream water. For a review of processes in the nitrogen and phosphorus cycles and a global inventory of storages and fluxes, see Pierrou (1976), Richey (1983) and Soderlund and Svensson (1976).

1 *Nitrogen sources and chemical transformations in soils*

Figure 2 illustrates the sources of dissolved and particulate nitrogen and phosphorus to streams. Figure 3 shows in more detail the specific processes responsible for the disposition of various forms of nitrogen in agricultural and forest soils. Most nitrogen in the litter and soil exists as organic nitrogen, which is unavailable to plants. Nitrogenous organic matter, however, is subject to chemical and biological degradation, thereby releasing ammonia (NH_3) in the mineralization process. Nevertheless, these same biological organisms simultaneously incorporate nitrogen into their plasma for growth in the process termed immobilization. Positive correlations have been found between mineralization in soils and soil moisture, temperature and the presence of soil animals and microorganisms (Wollum and Davey, 1975).

A second major process, nitrification, is responsible for the transformation of ammonia to nitrite then to nitrate. The second transformation is quick, such that nitrite is typically present in small amounts and for only short durations. The rate of nitrate supply through nitrification is dependent on the rate of mineralization, acidity of the soil, soil moisture and the cation-exchange capacity (CEC). Soils with moisture contents near saturation decrease the amount of oxygen available, thus inhibiting oxygen levels crucial to nitrification. Wollum and Davey (1975) pointed out:

> the effect of CEC on nitrification is at least two-fold. Under conditions of low CEC, there are fewer exchange sites for ammonium and, with less ammonium adsorbed, there will be a greater amount of ammonium in the soil solution. Also, the soil solution pH will be higher, with the tendency for a greater percentage of the reduced N to be in the form of ammonium.

Likens *et al.* (1969) noted that nitrification becomes an especially important factor controlling nitrogen releases from soils when these soils contain low amounts of

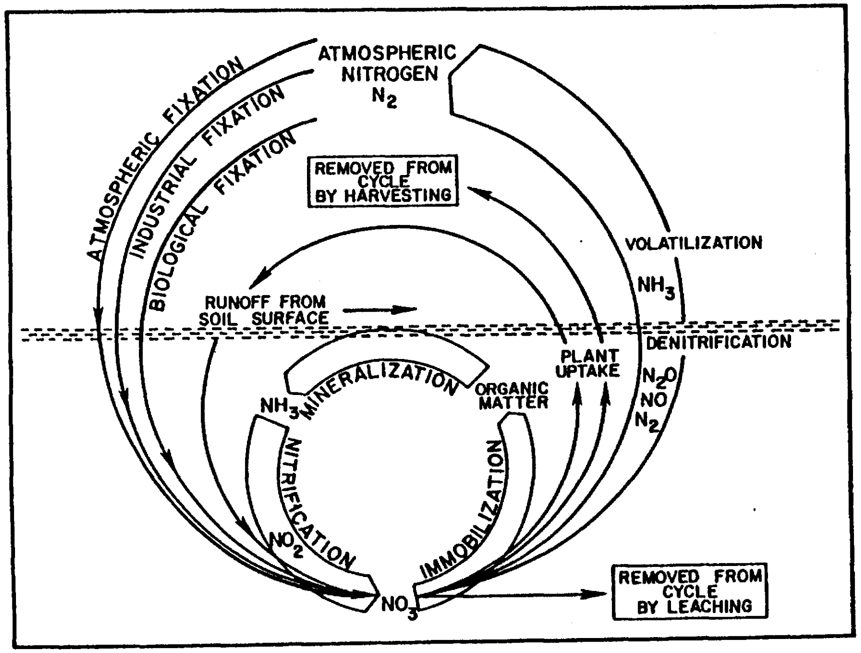

Figure 3 The nitrogen cycle (after Donigian and Crawford, 1976b).

clay and organic matter. Following deforestation, studies have shown that increased biological nitrification is responsible for the increased release of nitrate anions (Likens *et al.*, 1970). In acidic forest soils, nitrate concentrations in i:'reams relate directly to the rate of nitrification (Coats *et al.*, 1976).

2 *Phosphorus storage and chemical transformations in soils*

Geochemical weathering and mineralization are the two principa' processes accounting for the availability of the phosphate anion for leaching or adsorption (Figure 4). Organic phosphorus levels are generally high in surface soils due to accumulation in organic matter, and it is this fraction which is subject to mineralization (McElroy *et al.*, 1976). Inorganic phosphorus may exist in three forms in unfertilized soils: (1) as apatite, the calcium-phosphate mineral; (2) as an adsorbed ion on the surfaces of iron, aluminum and calcium soil colloids; and (3) as an ion present within the matrices of iron and aluminum compounds (Ryden *et al.*, 1973).

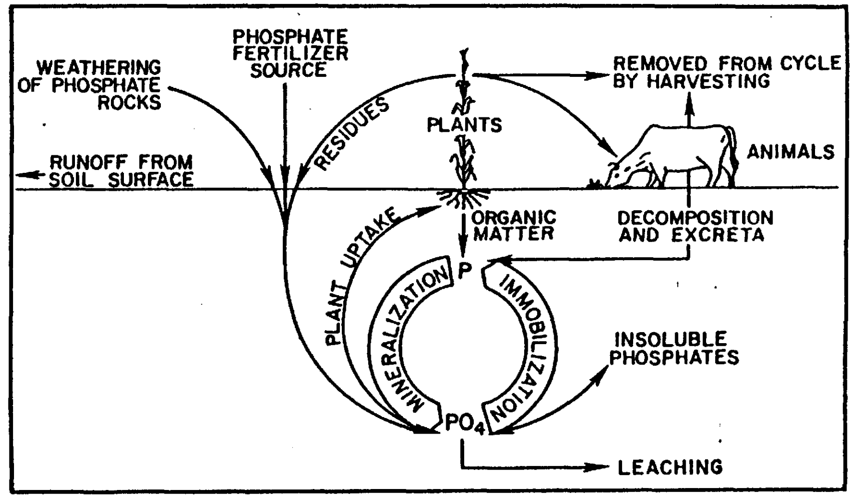

Figure 4 The phosphorus cycle (after Donigian and Crawford, 1976b).

3 *Adsorption and transport of nitrogen and phosphorus*

The supply of ammonia and nitrate in unfertilized soil depends primarily on the relative rates of mineralization and nitrification, while the supply of the orthophosphate anion depends on the rate of mineralization. When water comes into contact with the supplies of these nutrients in soil, these ions may either be adsorbed, desorbed or dissolved. Adsorption occurs when a hydrogen cation (H^+) or other non-nutrient cation is introduced to the soil along with a mobile (soluble) anion. With some major exceptions, clay-sized mineral particles and organic colloids typically have a high CEC, facilitated by the high surface area per unit volume and the negative electric charges of the clay particles.

There are two basic types of adsorption, specific and non-specific. During non-specific adsorption, ions are held in a diffuse double layer next to the charged colloid surface (Johnson and Cole, 1977). Ammonia and nitrate both exhibit non-specific adsorption which is dependent on soil acidity. A soil with a low pH (i.e., high H^+ concentration) will be positively charged, providing numerable exchange surfaces for anion adsorption. Thus, if the positive charge of soil is reduced by raising the pH, nitrate and ammonia can be desorbed (Hsu and Jackson, 1960). In specific adsorption, anions enter into coordination with the metal oxide by displacing another ion. In this case, the anions became bonded to two or more ions in the crystal structure.

The transport rates of these ions, and the proportion which remain dissolved, vary greatly among ammonia, nitrate and orthophosphate. The various ions have been ranked for their displacing capacity, an inverse indication of their relative mobility in forest soils. The order of ease in leaching is generally ranked: NO_3, $NO_2 > NH_4 >$ organic $N > PO_4$ (Brown, 1974; Johnson and Cole, 1977; Khanna, 1981). Consequently, nitrate is relatively mobile while the phosphate anion is tightly bound to exchange sites. Adding to the potential for nitrate to occur in the dissolved fraction is the fact that it is not subject to immobilization or mineralization as readily as ammonium. Although the ammonium ion is relatively mobile, it will still readily adsorb to exchange sites if the other cations are limited in supply (McElroy *et al.*, 1975; Wollum and Davey, 1975). Ammonium is not leached in large quantities unless a heavy rain follows applications of urea fertilizer. Moreover, NH_4^+ is easily nitrified, even during transport. Some ammonium is also taken up by plant roots and some released as NH_3^+ gas (Khanna, 1981). It should be emphasized that neither organic nor inorganic phosphorus are very soluble. When phosphorus occurs in dissolved form, it is readily removed by adsorption through plant roots or adsorption to soil particles (Cooper, 1969). A study in the dryland wheat region of eastern Washington found that more than 90 per cent of the phosphate exposed to sediment was absorbed (Carlile *et al.*, 1974). Phosphorus will be retained more in forest soils than in more xeric soils because of the greater fixation by iron and aluminum in the former and the greater solubility due to calcium in the latter (Hsu, 1965; Lajtha and Schlesinger, 1988; Novotny and Chesters, 1981; Vijayachandran and Harter, 1975)

The transport of nitrogen and phosphorus ions to the aquatic ecosystem is accomplished by two principal processes. Overland flow delivers both dissolved and particulate nutrients. Leaching, percolation and interflow delivers an added portion of dissolved nutrients to surface waters, contributing to stream concentrations during and long after storm events and accounting for nearly all base flow concentrations (Figure 2). Legg and Meisinger (1982) pointed out that dissolved losses of nitrogen are high when soil NO_3^- levels are high and downward movement of water is sufficient to remove NO_3 below the rooting depth. Because nitrate is relatively mobile within soil, it is easily dissolved and leached to a zone of deep-root penetration for plant uptake, if it does not encounter an impermeable horizon or is released to the stream. Meanwhile, ammonia and orthophosphate tend to accumulate in the forest floor litter or surface horizons of the soil, areas subject to soil particle detachment by raindrop splash and transport by overland flow (Corey and Biggar, 1969). The nutrient loading contribution of mass movement has not been addressed in the literature. This is a critical research gap considering the high toxicity of nitrite (NO_2) when applied in slug dosages; otherwise, nitrite is rarely considered as a nonpoint source pollutant in the literature. The comparative magnitude of the various nutrient transport processes depends on site-specific climatic and catchment factors controlling the dominance of overland flow and subsurface flow. The loss of nitrogen and phosphorus by transport of sediment with these adsorbed nutrients is a particular problem in light of the 'selective nature of the erosion process' described by Massay and Jackson (1952) and Singer and Rust (1975). Land surface erosion tends to first remove the organic-rich clay and silt-sized particles which account for the greatest concentrations of particulate nutrients.

Stevenson (1986) estimated that the total amount of nitrogen delivered to surface waters in the United States is 2 to 3.7 × 10^9 kg/y in dissolved form and 4.5 × 10^9 kg/y in particulate form. Omernick (1976; 1977) has developed empirical models and corresponding maps of nonpoint production of total nitrogen, total inorganic nitrogen and total phosphorus to surface waters of the 48 contiguous United States. However, he does not distinguish between the dissolved and particulate fractions. In his 1977 study, Omernick outlined a method for estimating concentrations of these nutrients from his maps that suffers a serious flaw. The maps illustrate units of equal concentration and the user is instructed to obtain a weighted mean concentration for any point in a catchment based on the relative areal extent of the various map units. The method he describes does not account for the contrast in water yield which will alter the nutrient yield from the various map units.

4 *Nitrogen and phosphorus in surface waters*

Once ammonia, nitrate and orthophosphate enter surface waters, they are subject to the same processes of adsorption, desorption and dissolution outlined earlier, as well as storage in alluvial materials. If a stream with a high concentration of dissolved phosphorus encounters an input of sediment with lower concentrations of adsorbed phosphorus, the sediment will adsorb additional phosphorus from the water, thereby lowering the concentration of dissolved phosphorus (Corey and Biggar, 1969). This effect is commonly observed with inputs of streambank sediment (Carlile *et al.*, 1974). Similarly, particulate phosphorus can be desorbed upon entering a stream with lower dissolved phosphate concentrations. Neither process for nitrogen ions has been reported in the literature. Desorption of phosphorus increases in direct proportion to the exchange surface area of the sediment. Because the ammonia, nitrate and phosphate ions adsorb to clay, silt and organic matter, the nutrients tend to remain in suspension longer, due to lower settling rates caused by the small-sized particles and the lower specific gravity of organic matter. Nevertheless, McKee *et al.* (1970) reported:

> a phosphorus reduction from 2000 ug/l to 150 ug/l as particulate matter settled from water passing through a 25-mi long reservoir that received 6.6 million tons of sediment annually. The particle size of the suspended sediment was very small and was comprised of 54 per cent clay, 40 per cent silt, and six per cent sand.

These nutrients may accumulate in reservoir or stream channel deposits if buried, with one study indicating no release of phosphorus from bog lake muds at depths greater than only 0.54 cm below the surface (Zieker *et al.*, 1956). In addition, Ryden *et al.* (1973) claimed that adsorbed phosphorus may become less exchangeable over time, culminating in a permanent removal of phosphorus from the stream system. Only through the activity of bottom-dwelling organisms or by turbulence will significant quantities of adsorbed nutrients be released (Cooper, 1969).

II Agricultural catchment studies of sediment-nutrient relationships

Numerous studies have examined the nutrient yield from agricultural lands (e.g see the summaries by Omernick, 1976; 1977; Uttormark *et al.*, 1974), but the distinction between dissolved and particulate export is not always clarified. The proportion of phosphorus delivered to streams by overland flow transport of sediment, as opposed to solution losses, may vary among study sites reflecting geographic variations of climatic and catchment properties. The studies of agricultural yields reviewed below were not exhaustive, but nevertheless illustrates the influence of selected variables on the dissolved and particulate flux of nitrogen and phosphorus (Table 1).

A five-year study by Burwell *et al.* (1974; 1976; 1977) on four catchments (30–61 ha) with well-drained, deep loess soils in southeastern Iowa revealed a major shift in the relative importance of particulate and solution transport depending on the cropping practice. The proportion of particulate nitrogen and phosphorus lost from catchments 3 and 4 were considerably less than from catchments 1 and 2 because of high sediment losses from the latter two which were contour-plowed. Catchments 3 and 4 were level-terraced. In addition, catchment 1 had heavier applications of fertilizer than catchment 2, while catchment 4 had heavier applications than catchment 3. The result for both sets of comparisons was that a greater percentage of nitrogen and phosphorus was released by particulate transport. The studies found that nitrogen losses will be minimized by using 'fertilizer application rates that do not exceed crop needs and using conservation practices that jointly minimize soil erosion and deep percolation' (hence optimizing plant uptake).

Kissel *et al.* (1976) examined nitrate losses on two catchments (4 ha each) with poorly drained clay soils on the Blackland Prairie of Texas. From a five-year study, the authors claimed that the long-term loss of particulate nitrogen is 5.1 kg/ha/yr, or only 20 per cent of the total nitrogen loss, with organic nitrogen as the largest component. The authors also found that the loss of nitrate in runoff varied depending on the sequence of land-management practices before storm events. Also, the concentration of nitrogen in sediment could not be correlated with any runoff or catchment parameter.

A study on the Black Creek catchment in Indiana (unreported size) noted that between 11 and 23 per cent of total nitrogen losses were by particulate transport, with 52 to 70 per cent of phosphorus losses by particulate transport (Sommers, 1975). With an increase in fertilizer application, absolute amounts of particulate phosphorus losses increased, but no appreciable increases were reported for particulate nitrogen. Apparently, the nitrogen component in the fertilizer was sufficiently removed by plant uptake or any excess was quickly nitrified to nitrate and lost to subsurface flow.

Two studies are focussed on the effect of cropping practices on the disposition on nitrogen and phosphorus. McDowell and McGregor (1980) measured the dissolved and particulate losses of nitrogen and phosphorus on highly erodible loess soils in northern Mississippi subject to different cropping practices. With conventional tilling of soybeans, high rates of erosion were observed, accounting for 99 per cent of the total nitrogen losses and 100 per cent of the total phosphorus losses. With no-till farming of soybeans and soybeans rotated with either wheat or corn, the particulate losses ranged between 11 and 50 per cent of nitrogen losses and between 40 to 81 per cent of phosphorus losses. Baker and Laflen (1983) found only minor changes in the dissolved and particulate loads from soybean fields after harvest when those fields had been fertilized in the fall by different methods. The particulate export ranged from 38 to 57 per cent for ammonium and from 78 to 97 per cent for phosphorus. Whether the fertilizer had been applied by injection or with or without tillage did not account for major differences. Similarly, chisel plowing or double disking did not influence the ratio of dissolved and particulate losses. However, artificial rainfall events were used to generate the nutrient export and these experiments may not approximate the actual timing, antecedent moisture conditions or intensity of rainfall events in the region. The more severe types of soil disturbance with chisel plowing and double disking will only accelerate particulate losses if rainfall energy is sufficient to generate overland flow exceeding the threshold of land surface erosion.

Several studies on agricultural catchments have only examined the relative losses of dissolved and particulate nutrients in overland flow, disregarding subsurface contributions. Johnson *et al.* (1976) used a catchment (330 km^2) in central New York with acid soils derived from glacial till to compare contributions of phosphorus from geochemical weathering, agricultural inputs and urban point sources. These sources release the majority of phosphorus during runoff events of short duration and high intensity, although the proportion of phosphorus lost by adsorption on sediment during storms was found to be constant. Most importantly, particulate phosphorus accounted for 78 per cent of the total phosphorus exported. A second study, by Olness *et al.* (1975) on seven cropland and four rangeland catchments (5–18 ha) in central Oklahoma, found particulate phosphorus losses accounting for 67 to 88 per cent of losses on the cropland and 91 to 97 per cent on the rangeland sites. A study by Schuman *et al.* (1973) on four small agricultural catchments in southwestern Iowa measured the nitrogen losses in surface runoff for three years. The authors found that particulate nitrogen losses accounted for 92 per cent of the total nitrogen loss, far exceeding values reported in other studies (Table 1). However, recall that these studies did not look at the contribution from subsurface flow which is probably major for nitrogen losses from these agricultural catchments.

A study by Romkens *et al.* (1973) on 85 km^2 plots with silt loam soils, which

Table 1 Particulate and dissolved losses of nitrogen and phosphorus from agricultural catchments

Location	Nitrogen ka/ha*	Nitrogen %Particulate	Nitrogen %Dissolved	Phosphorus kg/ha*	Phosphorus %Particulate	Phosphorus %Dissolved	Reference
Southeast Iowa							Burwell *et al.*, 1974; 1976; 1977
Wtsd #1	8.23	55	45	0.156	80	20	
Wtsd #2	5.00	71	29	0.099	74	26	
Wtsd #3	2.31	14	86	0.044	29	71	
Wtsd #4	7.33	17	83	0.081	41	59	
Delaware							Haith and Shoemaker, 1987
Corn	80.6	51	49	23.5	76	24	
Hay	47.4	19	81	5.9	64	36	
Pasture	14.8	31	69	2.8	71	29	
Inactive	7.4	47	53	1.9	84	16	
Barn yards	3.7	0	100	0.6	0	100	
Total USA							Hauck and Tanji, 1982
1930	0.23	55	45	–	–	–	
1947	0.25	57	43	–	–	–	
1967	0.27	60	40	–	–	–	
New York							Johnson *et al.*, 1976
Pasture and Inactive	–	–	–	13 360	78	22	
Mississippi							McDowell and McGregor, 1980
Till/soybeans	31.2	99	1	13.5	100	0	
No till/soybeans	0.98	39	61	1.12	17	83	
No till/soy-wheat	0.11	27	73	0.05	40	60	
No till/soy-corn	3.39	50	50	1.45	81	19	
No till/corn-soy	11.8	11	89	1.80	46	54	
Oklahoma							Olness *et al.*, 1975
Cotton/Dry.	–	–	–	5.01	88	12	
Cotton/Irr.	–	–	–	11.15	85	15	
Wheat	–	–	–	2.94	85	15	
Alfalfa	–	–	–	2.48	67	13	
Grazed #1	–	–	–	1.27	91	3	
Grazed #2	–	–	–	4.60	97	3	
Southwest Iowa							Schuman *et al.*, 1973
Wtsd #1	39.6	92	8	–	–	–	
Wtsd #2	25.1	92	8	–	–	–	
Wtsd #3	2.36	51	49	–	–	–	
Wtsd #4	3.04	86	14	–	–	–	

*Not normalized by time unit

used a rainwater simulator, produced a curvilinear relationship between the amount of sediment loss in overland flow and the amount of particulate nitrogen and phosphorus loss, for both fertilized and unfertilized situations. Such a relationship suggests that modelling efforts based on the sediment-nutrient correlation might be valid, except for the fact that these studies have ignored the contributions of nitrogen and phosphorus from subsurface flow. The relative losses by particulate and solution transport differ, depending on whether one is analysing storm runoff or base flow conditions.

The studies of sediment-nutrient relationships on agricultural catchments support the following conclusions:
1) Overland flow is a common occurrence, contributing substantial amounts of particulate nitrogen and phosphorus when soil conservation practices are poor;
2) with heavier fertilizer applications, a greater proportion of phosphorus was transported by particulate matter, but conflicting findings support no such conclusion for nitrogen; and
3) the release of nitrogen and phosphorus depends on the sequence of cropland management practices preceding storm events.

III Forested catchment studies of sediment-nutrient relationships

The studies of sediment-nutrient relationships cited for agricultural catchments emphasize the importance of particulate nutrient runoff, especially for phosphorus. This contribution results largely from overland flow, a process rarely observed on undisturbed forested catchments, except during periods wth frozen soils or in deciduous forests during long-duration storm events. An increase in land surface erosion following timber harvest is a topic widely reported in the literature, but the expected concurrent increase in particulate nutrient discharge has not been adequately discussed. Livingstone (1963) pointed out the danger in pursuing geochemical investigations which only account for the dissolved component of nutrient losses. For example, he suggests that phosphorus may exist at a level too low to detect in an inorganic solution, causing one to erroneously conclude that phosphorus is not present. It has already been shown that phosphorus in solution is readily transferred to sediment in adsorbed form if particulate phosphorus concentrations are lower than dissolved concentrations.

Similar misrepresentation can occur in studies of nitrogen loss from forested catchments, typified by results reported for nitrogen export from the Alsea River Experimental Catchments in the Oregon Coast Range (G. Brown, *et al.*, 1973). This study utilized three catchments, all with deep clay loam or shallow, stony, loam soils. Flynn Creek served as the control catchment with alder along the steambanks and Douglas fir on the hillslopes. Deer Creek had the same cover, but was 25 per cent clearcut in three small patches with the streamside alder left as a buffer strip. Needle Branch was entirely covered with Douglas fir but was clearcut, followed by a severe slash burn (Brown, 1974). The results reported by G. Brown *et al.* (1973) indicate that there was no significant change in the yield of nitrate after logging Dear Creek relative to observed values on Flynn Creek. The nitrate yield for the two years prior to treatment was 31 kg/ha and 25 kg/ha compared with the 24 kg/ha and 28 kg/ha for the two years after treatment. Moreover, the highest post-treatment concentration was only 2.96 mg/l, well below pretreatment levels. On Needle Branch, the yields increased from 4 kg/ha/yr (two-year average) to 15.5 kg/ha/yr, but still with a post-treatment concentration of only 2.10 mg/l.

The key point of these results is that only dissolved losses were reported, a fact not clarified later in a widely quoted publication by Brown (1974). This is particularly significant when one considers the marked increase in sediment yield for Deer Creek and Needle Branch following timber harvest, increasing from 780 kg/ha/yr to 1800 kg/ha/yr for the former, and from 390 kg/ha/yr to 1780 kg/ha/yr for the latter. Thus, the reported nitrate losses do not include the proportion adsorbed on sediment and subsequently lost by land surface erosion, mass wasting, and channel erosion. The authors admitted that their results account for less than one half of the total nutrient loss because of this effect (Brown, G. *et al.*, 1973). This supposition was based on results from a forested catchment study in a contrasting physiographic setting. The authors also reported that sediment loss began to recover by the third year after treatment, suggesting that nutrient release would likewise decrease from particulate transport.

A more complete description of sediment-nutrient relationships was reported by Fredriksen (1971) for the H.J. Andrews Experimental Forest in the western Oregon Cascades. The soils in the study area are deep clays (with a high CEC), containing a high amount of organic material well distributed throughout the soil. High amounts of precipitation occur, predominantly in the winter in the form of rain. A Douglas fir forest was clearcut and burned in 1966. Fredriksen analysed annual losses of chemical constituents on inorganic and organic particulate matter as well as the dissolved component for two years following the treatment.

Fredriksen found that greater than 50 per cent of the total nitrogen enters the stream either combined with organic particulates or adsorbed to inorganic particulates. This was attributed to the extensive surface erosion which diminished in the second year after treatment. Meanwhile, all nitrogen losses in the control catchment were in solution. Consequently, the proportion of nitrogen transported with particulate matter also declined. Fredricksen claims that controlling soil erosion will also mitigate the nutrient release. Moreover, the dissolved nitrate losses could also be retarded by avoiding slash burning. Fredricksen does not state that avoiding slash burning will also mitigate particulate nutrient losses by preserving the protective vegetative cover over mineral soil, thereby controlling sediment losses. Fredricksen concludes that nutrient transport by adsorption to inorganic sediment and in combination with organic matter is much less prevalent than dissolved losses in undisturbed forest catchments, because of the minor importance of land surface erosion.

An understanding of contrasting catchment and climatic controls on nutrient sources, movement and adsorption comes from work in the Hubbard-Brook Experimental Forest (Likens *et al.*, 1970). The soils in the study area are shallow, low in clay content (low CEC), are generally unfrozen, and have low amounts of organic matter which is most concentrated in a distinct surface layer. The study area has moderate amounts of precipitation, evenly distributed throughout the year, with 50 per cent occurring as snow. The net result is that interflow is much greater than deep percolation (Bormann *et al.*, 1969). The birch/beech/maple forest was clearcut and herbicides applied to control vegetation growth for two years following the clearcut. Under these conditions, one would expect adsorption of ions to sediment to be minimized and leaching to be of greater importance. Stream water quality was analysed for particulate and dissolved nitrogen and phosphorus among other constituents.

As expected, particulate losses accounted for only three per cent of the total nitrogen export and 63 per cent of the phosphorus export (Likens *et al.*, 1977). The authors point out that the concentrations of dissolved nutrients are not affected by flow rates, but they are related to the annual water yield. On the other hand, particulate losses were shown to relate directly to flow rates. Annual streamflow increased 39 per cent during the first year after treatment and 28 per cent during the second year above expected values had deforestation not occurred. Thus, timber harvest was directly related to an increased proportion of particulate nutrient losses. Most of this increase was observed for inorganic particulates rather than the organic fraction (Likens *et al.*, 1970). The greatest seasonal discharge of particulates occurred during the summer, with most of the annual load moved during storms. Hobbie (1973) added that the inorganic particulate phosphorus lost to bedload increased 12 times over that experienced in the catchment before deforestation. Haith and Shoemaker (1987) found the same relative disposition of nitrogen but an inverted pattern for phosphorus in their modelling study of the West Delaware River watershed. Currier and O'Hayre (1980) note that the forest treatment was extreme in the Hubbard Brook study, and they cite 16 other studies that point to minor increases following treatment. Unfortunately, the data for these 16 studies do not separate the export of nitrogen and phosphorus into dissolved and particulate fractions.

Singer and Rust (1975) presented findings which include the influence of frequent periods of frozen soils on nutrient discharge in an oak-basswood-maple forest in Minnesota. Ignoring subsurface losses throughout the year, claimed to be insignificant, the authors found that 33 per cent of the phosphorus was lost by particulate transport and the remaining 67 per cent occurred as dissolved losses. The major loss occurred during the spring snowmelt when the soil was frozen, at which time both the particulate and dissolved components increased (Table 2).

A study by Crisp (1966) in the Rough Sike catchment in England reported unusually high particulate losses of both nitrogen and phosphorus. However, it must be noted that bog vegetation was the dominant land cover and peat erosion contributed the particulate forms of nitrogen and phosphorus.

There are many other studies of nutrient loss from forested catchments, but

Table 2 Particulate and dissolved losses of nitrogen and phosphorus from forest catchments

	Nitrogen			Phosphorus			
Location	ka/ha*	%Particulate	%Dissolved	kg/ha*	%Particulate	%Dissolved	Reference
England							
Bog vegetation	17.6	83	17	0.84	53	47	Crisp, 1966
Oregon							Fredriksen, 1971
1967 Cut	0.98	63	37	–	–	–	
1967 Control	0.03	74	26	–	–	–	
1968 Cut	0.71	44	56	–	–	–	
1968 Control	0.03	95	5	–	–	–	
Delaware Forest	63.8	3	97	2.55	33	67	Haith and Shoemaker, 1987
New Hampshire Wtsd #6	4.01	3	97	0.019	63	37	Likens *et al.*, 1970; 1977
Minnesota Mixed forest	–	–	–	0.09	33	67	Singer and Rust, 1975

*Not normalized by unit time

the distinction is rarely made between particulate and dissolved losses. Ryden *et al.* (1973) cited six studies from forested catchments over a wide geographic range, noting that phosphorus losses range from 0.02 and 0.68 kg/ha/yr, with dissolved inorganic losses accounting for 0.02 to 0.07 kg/ha/yr. By ignoring dissolved organic phosphate losses, comparisons of dissolved versus particulate losses are prevented. A study on 25 coniferous and subalpine forested catchments (3–141 km^2) in the Lake Tahoe-Truckee River drainage included a sampling programme for suspended sediment, nitrate, organic nitrogen and dissolved phosphorus. This study outlines the sources of nitrogen and phosphorus, their movement through the system, and the processes responsible for removing these nutrients from solution (Brown, J. *et al.*, 1973). Unfortunately, the authors did not quantify the relative proportions of particulate and dissolved losses. A study by Timmons *et al.* (1977) in an aspen/birch forest in northern Minnesota claimed that twice as much ammonia was lost as nitrate in both surface and subsurface flow, but the distinction between particulate and dissolved losses is not clear. A study by the Chesapeake Bay Center for Environmental Studies (1977) on the Rhode River Experimental Catchment (59 per cent forested) found no significant correlation between the volume of sediment lost the nutrient concentrations in runoff. Kovda (1971) and Likens *et al.* (1977) cited dozens of studies that quantify nitrogen and phosphorus yield from a variety of forest ecosystems worldwide, but few report the relative proportion lost in particulate and dissolved form.

It appears that the degree to which silvicultural activities increase overland flow will have a direct bearing on the proportion of nitrogen and phosphorus which is transported by particulate matter. The effect of fertilization during forest regeneration on the relative importance of the dominant nutrient transport processes is not well understood, but from the studies on agricultural catchments it may be hypothesized that a greater proportion of phosphorus will be transported by particulate matter with uncertain results for nitrogen. Ideally, fertilization should be timed to occur during periods when overland flow will be minimized. The studies of sediment-nutrient relationships on forested catchments support the following conclusions:

1) The proportion of nitrogen and phosphorus which are lost by particulate transport following timber harvest depends on site-specific catchment and climatic factors summarized by Morisot (1981), including soil depth, clay content (CEC), seasonality of precipitation and runoff, the occurrence of frozen soils, storm period rainfall energy and the importance of snowmelt runoff;

2) sediment control practices will also mitigate the loss of particulate nutrients; and

3) the lack of studies in forested catchments which make the distinction between dissolved and particulate losses makes it difficult to justify the general conclusion often presented in the literature that timber harvest does not increase stream nutrient levels.

IV Nonpoint sources modelling of sediment-nutrient relationships

The US Environmental Protection Agency (EPA) has sponsored at least two nonpoint source models for nitrogen and phosphorus loading of streams which depend on the sediment-nutrient relationship. As required by Section 3–4 of the 1972 Amendments to the Federal Water Pollution Control Act (PL 92–500), the EPA is to issue guidelines for the identification and evaluation of the nature and extent of nonpoint sources. One model developed for this purpose is the loading function of nitrogen and phosphorus derived by the Midwest Research Institute (MRI) through EPA funding (McElroy *et al.*, 1976). The particulate yield of nitrogen, $Y(NT)_e$, or phosphorus, $Y(PT)_e$, from any landuse category is predicted by multiplying sediment yields, $Y(S)_e$, by the concentration of total nitrogen, $C_s(NT)$, or phosphorus, $C_s(PT)$, in the soil and by an enrichment factor, r_n or r_p. The enrichment value is determined empirically as the concentration of nitrogen or phosphorus in the eroded material divided by its concentration in the soil. The enrichment ratio for nitrogen, r_n, generally ranges in value from 2 to 5; the phosphorus enrichment ratio, r_p, generally ranges in value from 1 to 3. A dimensional constant, a, is included with a value of 10 if SI units are used (20 for English units). The sediment yield, $Y(S)_e$, is estimated first by the Universal Soil Loss Equation with consideration given to the sediment delivery ratio, S_d. The equations take the form:

$$Y(NT)_e = a \cdot Y(S)_e \cdot C_s(NT) \cdot r_n \qquad (1)$$

$$Y(PT) = a \cdot Y(S)_e \cdot C_s(PT) \cdot r_p \qquad (2)$$

The authors claimed that these loading equations do not include dissolved losses and conclude that the erosion-based loading function will give inaccurate estimates for nitrogen discharge from forested catchments which have a minimum amount of soil erosion. The authors otherwise assume 'that the nutrients . . . are carried through surface runoff and that most of these are removed with sediment'.

The problems with the MRI nutrient loading functions are manifold, especially with regard to use in forested catchments. There are many difficulties associated with evaluating each factor in the Universal Soil Loss Equation. Moreover, its applicability is limited to areas with slopes less than 20°, uncommon to many forested catchments, particularly in the Pacific Northwest. Secondly, the studies cited earlier regarding sediment-nutrient relationships on agricultural catchments do not support the notion that 'phosphorus is carried almost entirely on sediment', as claimed in the MRI report. The proportion of phosphorus or nitrogen which undergoes particulate transport as opposed to transport in solution depends on the sequence of crop-management practices before storm events as well as the amount of fertilization (Table 1). Indeed, the MRI loading function is not applicable to forested catchments because of the significant proportion of nitrogen and phosphorus which is lost by leaching, percolation and interflow, even after timber harvest. Nevertheless, the EPA promotes this model for use 'with caution' for estimating the insoluble component (Currier and O'Hayre, 1980).

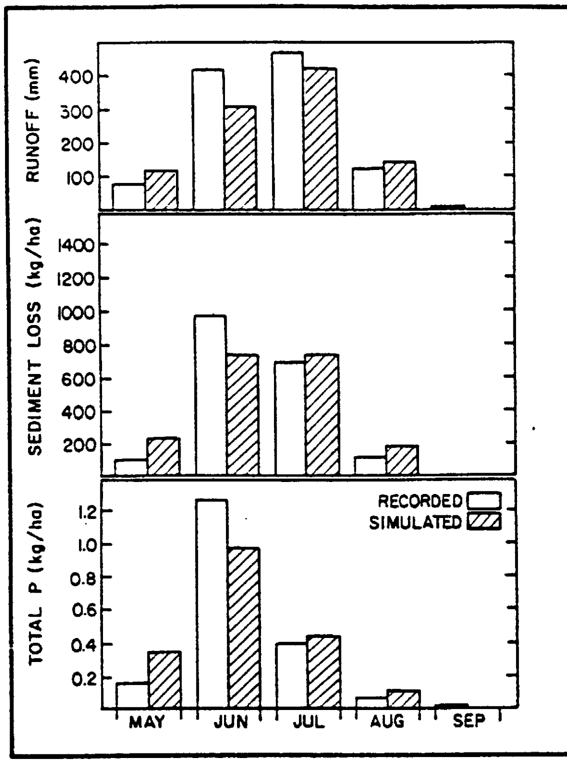

Figure 5 Monthly runoff, sediment and total phosphorus loss from a small experimental agricultural catchment in Georgia used for testing the Hydrocomp model by Donigian and Crawford (1976a; 1976b; 1977).

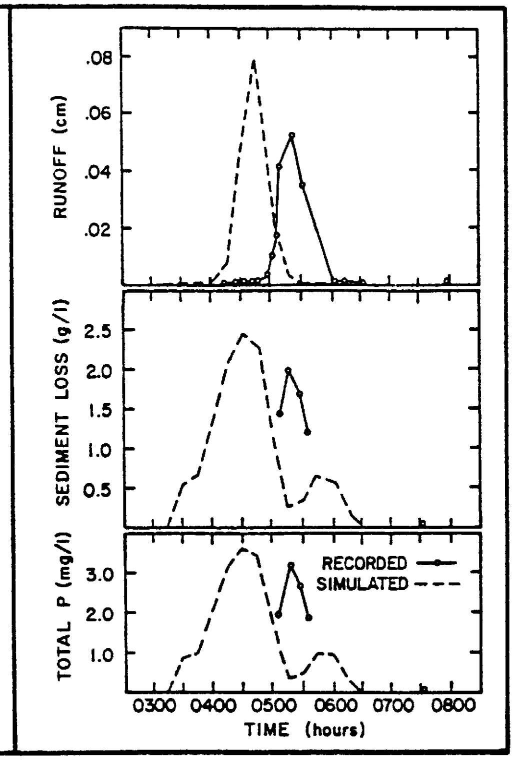

Figure 6 Runoff, sediment loss and total phosphorus concentrations for the same catchment as in Figure 5 for the storm of 23 May 1974 (after Donigian and Crawford, 1977).

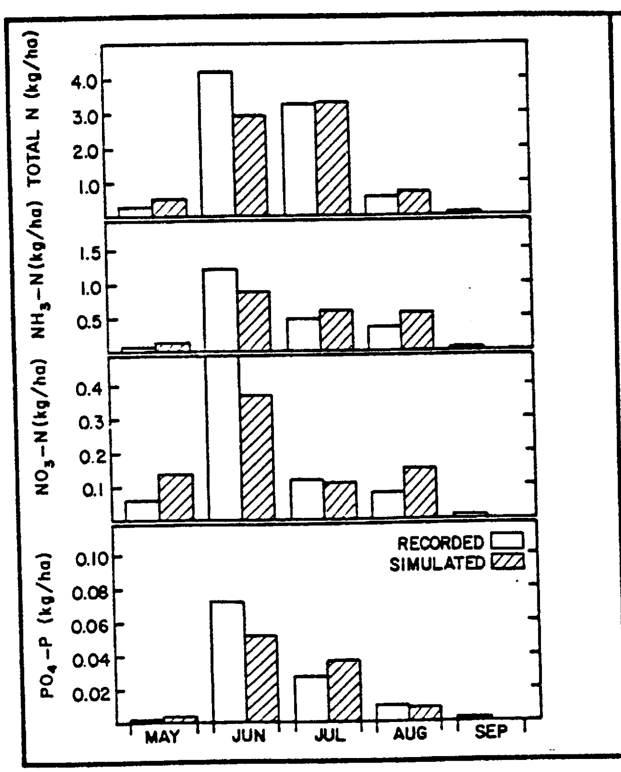

Figure 7 Monthly total nitrogen, ammonia, nitrate and phosphate from the same catchment as in Figure 5. Values expressed are for the May-September period, 1974 (after Donigian and Crawford, 1977).

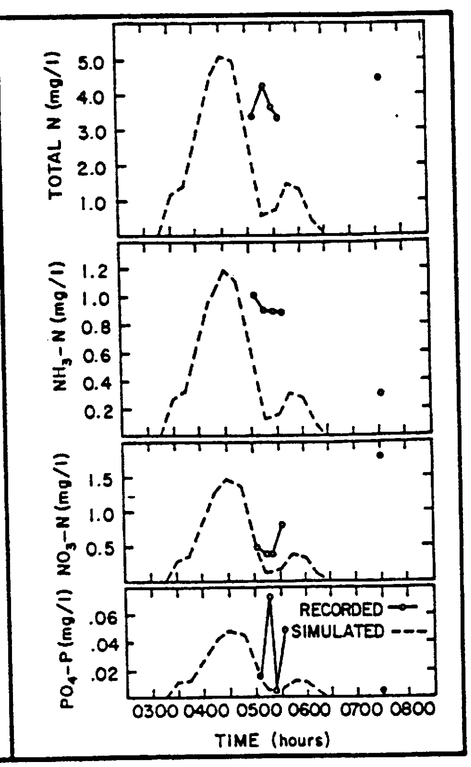

Figure 8 Total nitrogen, ammonia, nitrate and phosphate concentrations for the same catchment as in Figure 5 for the storm of 23 May 1974 (after Donigian and Crawford, 1977).

Hydrocomp Incorporated developed a second nonpoint source model for the EPA which utilizes the sediment-nutrient relationship (Donigian and Crawford, 1976a; 1976b; 1977). The Hydrocomp Model used computer simulation of hydrologic algorithms to calculate nitrogen and phosphorus loads associated with sediment in surface runoff from urban and agricultural catchments. The various hydrologic parameters include interception, impervious area, infiltration, interflow, soil moisture, overland flow, evapotranspiration and snow heat transfer. Although the Hydrocomp Model is far more complex than the loading functions derived by MRI, the Hydrocomp model still depends on the degree to which sediment can be used as an indicator of nutrient levels. The Hydrocomp Model, therefore, does not address dissolved losses but quantifies only surface runoff since the model was originally derived for sediment releases alone. In other words, the success of the Hydrocomp Model is predicated on the explanation of the majority of nitrogen and phosphorus discharge by particulate transport.

Johnson *et al.* (1976), Olness *et al.* (1975) and Schuman *et al.* (1973) considered only overland flow in concluding that a majority of total phosphorus losses (67 to 97 per cent) and total nitrogen losses (92 per cent) could be explained by particulate transport from agricultural catchments (Table 1). Dissolved losses by subsurface flow were not examined. For this reason, Donigian and Crawford (1977) claimed that the literature supports the results from testing the Hydrocomp Model, indicating that total loads of these two nutrients can be reasonably simulated (Figures 5–8). The peak values are within 20 per cent for storm concentrations and monthly yields of total nitrogen and total phosphorus, but the rising and falling limbs are out-of-phase between the recorded and simulated curves. Monthly yields were also simulated fairly accurately for ammonia, nitrate and phosphate ions (Figures 5 and 7), but there was a wide discrepancy in the timing and magnitude of storm peak values between the recorded and simulated curves (Figures 6 and 8). Surprisingly, the authors attributed the discrepancy for phosphate to dissolved losses, claiming that the proportion lost by this process is similar to that for nitrate. This is in direct contrast to the studies cited earlier which emphasized the importance of particulate transport of the phosphate anion, related to its strong displacing capacity.

The authors concluded that the Hydrocomp Model can simulate nutrient loadings only in areas where subsurface contributions are minimal. This implies that the application of the Hydrocomp Model to forested catchments is limited, except for calculating monthly yields following timber harvest. Before modelling nitrogen and phosphorus loads with sediment as an indicator, subsurface nutrient discharge must be incorporated as well as dissolved and particulate transport in overland flow. In this manner, the proper proportion of the total load transported by sediment can be calculated for various climatic and catchment conditions. A promising effort in this direction is the set of 'Generalized Watershed Loading Functions' proposed by Haith and Shoemaker (1987) which separates the dissolved fractions of nitrogen and phosphorus for a variety of landuse settings.

V Conclusion

Ammonia (NH_3), nitrate (NO_3) and orthophosphate (PO_4) are the principal nonpoint source forms of nitrogen and phosphorus. They may be released to surface waters by dissolved transport in subsurface water or by dissolved and particulate transport in overland flow. Particulate transport includes the adsorption of these ions to inorganic particles as well as the adsorption to, or combination with, organic materials. Nitrate has a low displacing capacity and is consequently very mobile, being subject to dissolved losses. Phosphate has a strong displacing capacity causing it to be readily adsorbed on sediment. Ammonia tends to accumulate in the forest floor litter or upper soil and may be adsorbed as well as dissolved before mineralization.

Studies of sediment-nutrient relationships on agricultural catchments revealed that overland flow is a common occurrence, contributing substantial amounts of particulate nitrogen and phosphorus when soil conservation practices are poor. The release of nitrogen and phosphorus depends on the sequence of cropland management practices preceeding storm events.

The distinction between dissolved and particulate nutrient transport is rarely reported in the literature for forested catchments. Studies in the Hubbard-Brook Experimental Forest and H.J. Andrews Experimental Forest present contrasting results regarding the percentage of phosphorus transported by sediment following timber harvest. These differences can be attributed to regional differences in the forest soils, specifically soil depth, content of clays and organic matter (i.e., CEC) and the rate of nitrification. Studies of nutrient release from forested catchments which do not account for the particulate component may provide misleading results. Because streambank erosion is dominant in undisturbed forests, the phosphorus-scavenging effect of this sediment should be investigated because of the typical low concentrations of phosphorus in the streambank sediment and the subsequent adsorption of dissolved phosphate. The results summarized for agricultural catchments above may also apply to forested catchments.

Thus, a review of sediment-nutrient studies on agricultural and forested catchments reveals no constant proportion of nitrogen or phosphorus transported by either sediment or solution, although trends are apparent. This observation undermines the usefulness of any nonpoint source models for nitrogen and phosphorus discharge, especially from forested catchments, unless the models can account for synoptic climatic conditions (e.g. frozen soils, storm rainfall energy, snowmelt), the geographic variability in catchment controls on the sources, adsorption, and movement of these ions and the effect of contrasting cropping patterns and conservation practices. Nitrogen and phosphorus export also changes over time as a part of ecosystem succession (Reiners, 1981), another factor not incorporated into nonpoint source modelling. Before sediment can be used as an indicator of nitrogen and phosphorus loads from forested catchments, the transport of these nutrients in subsurface and overland flow must be quantified, with distinction between the dissolved and particulate component.

The proportion of nutrients transported by sediment can then be related to observed levels of the various ions to develop a factor quantifying the importance of this transport process depending on catchment factors, synoptic climatology, and the sequence of silvicultural practices.

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