Recent Developments in the Explanation and Prediction of Erosion and Sediment Yield (Proceedings of the Exeter Symposium, July 1982). IAHS Publ. no. 137.

Influence of season, source and distance on physical and chemical properties of suspended sediment

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ABSTRACT Understanding of source, pathways and fate of suspended sediment-associated nutrients and contaminants is now recognized as an important element of water resource management. The geochemically active fraction (<62 µm) of the suspended sediment load concentrates nutrients, metals and organic contaminants by several orders of magnitude above ambient water concentrations. Because this sediment fraction tends to be flow dependent, is not depth dependent nor adequately characterized by bottom sediments, collection of suspended sediment for geochemical purposes requires new sampling strategies. Mineralogy and major element analysis together with systematic seasonal changes in physical characteristics of suspended matter can assist in identifying seasonally variant source areas. Changes in sediment chemistry during downstream transport reflect a suite of biogeochemical processes which should be explicitly considered when assessing the environmental relevance of sediment quality.

INTRODUCTION

Of growing significance in the field of water resources is the role of suspended sediment (mineral + organic) in the conveyance of nutrients and contaminants across eroding land surfaces and into and through aquatic systems. Conventional research into erosion and sedimentation typically deals with the coarse-grained (>62 µm) fraction. This fraction is, however, of marginal significance in water or sediment quality issues. The linkage between sediment sources, sediment conveyance and water quality concerns such as eutrophication and contaminant transport, is centred upon the physi physical and biogeochemical behaviour of the fine-grained (wash load = <62 µm silt/clay + organic solids) fraction.

The role of suspended sediment in the transport and biogeochemical cycling of nutrients and contaminants in aquatic systems is now well established in the literature (Allan, 1979; Förstner & Whittmann, 1981; Shear & Watson, 1977; Ongley *et al.*, 1981). It is now beginning to be accepted that, for water quality purposes, suspended sediment must be explicitly examined rather than merely included within "total" analyses of water samples. The physical and geochemical dynamics of sediment transport, especially in terms of biogeochemical cycling of sediment-associated nutrients and contaminants is not, however, well understood.

This paper explores a variety of issues pertinent to suspended solids within fluvial systems. In particular, the implications for sampling, and the influence of source, seasonality and distance upon geochemical characteristics are discussed. Most of the examples are drawn from ongoing research programmes of the author.

CHEMICAL SIGNIFICANCE OF SUSPENDED SEDIMENT

The environmental relevance of suspended sediment lies in the chemically reactive nature of fine-grained solids. Clay minerals, because of large surface area, tend to have high adsorptive capacities for cations and anions. Manganese and iron oxides which commonly coat clay minerals adsorb certain metals more effectively than fresh clay surfaces. A general sequence of sorption capacity for metals by fine-grained solids is that of Guy *et al.* (1975):

MnO_2 > humic acids > hydrous iron oxides > clay

Although fine-grained suspended sediment is here considered to include all material <62 μm in equivalent spherical diameter, Fig.1 illustrates that it is the fine end of the particle-size spectrum which is of primary importance. There are, however, many unresolved questions about the effective size of particles in natural fluvial systems arising from, for example, flocculation (e.g. Logan, 1978).

Suspended sediment includes organic in addition to mineral material. Although overlooked by sedimentologists and geomorphologists, organic matter is probably more relevant in biogeochemical cycling of nutrient and contaminants than mineral material (Jonasson, 1977; Gardner, 1974). Organic sediment can include organic coatings on mineral sediment, discrete organic debris (ignoring larger material such as floating weeds, etc.), and a variety of organic substances ranging downwards from small organic particles through colloids to organic acids. The role of organic colloids and humic substances in sorption/desorption and chelation processes is beyond the scope of this paper. Conventional measures of organic contribution to suspended sediment load are summarized in Ongley *et al.* (1981) and range up to 69% for summer/fall periods (Pierce & Dulong, 1977).

The geochemical significance of suspended sediment relative to solute chemical forms has been reported by many authors. It should be stressed, however, that there is much less known about the biological relevance (e.g. bioavailability) of sediment chemistry. Biological relevance must be the final determinant of the environmental significance of suspended matter.

Nutrients

Unlike nitrogen, phosphorus is primarily associated with solid matter. The enrichment process during conveyance across agricultural surfaces is well known to soil scientists (Fig.1). Less well known is the important role suspended sediment plays in removing soluble orthophosphate (which is biologically available)

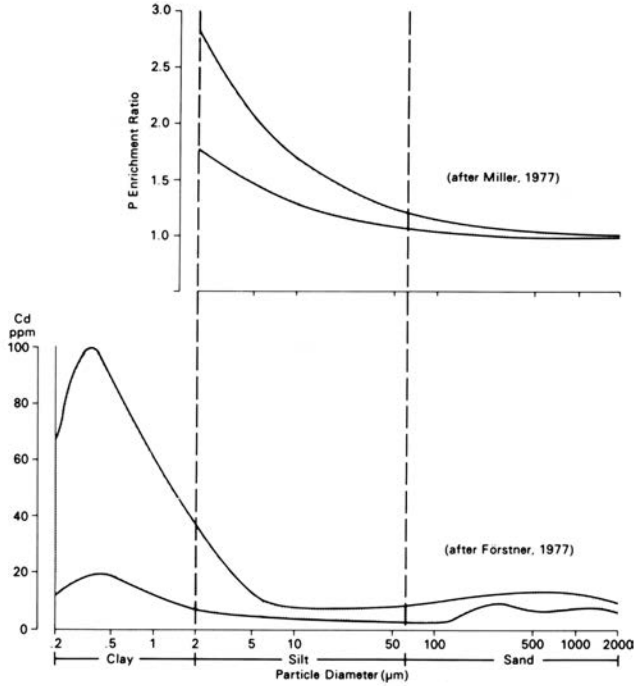


FIG.1 Phosphorus enrichment and cadmium concentration as a function of particle size.

from solution (Logan, 1978). The proportion of sediment-associated phosphorus to total-P flux on an annual basis in a variety of fluvial settings is reported to range from 78 to 95% (Johnson *et al.*, 1976; Lake Erie Wastewater Management Study (LEWMS), 1975; Logan, 1978; O'Neill, 1979; Rigler, 1979). Although there is considerable controversy over the bioavailability of sediment-associated phosphorus, data from chemical fractionation (e.g. Williams *et al.*, 1976; Logan *et al.*, 1979) and algal assay (Golterman, 1977; Williams *et al.*, 1980), suggest that a significant proportion of sediment-associated phosphorus is potentially available to biota and must, therefore, be taken into account when developing monitoring strategies and in formulating nutrient abatement programmes for point and diffuse sources.

Metals and contaminants

The quantitative significance of suspended sediment in heavy metal transport in fluvial systems is well established (Förstner & Salomons, 1980). The reader is referred to Förstner & Whittman (1981) for a comprehensive review of metals in the aquatic environment.

Many organic contaminants of toxicological importance such as nonionic organochlorine pesticides, PCB, mirex and the lethal 2, 3, 7, 8 TCDD isomer of dioxin, have low to nil solubility and are found almost exclusively in association with solid matter (including bioaccumulation in living tissue).

One must conclude that the geochemical properties of suspended sediment have profound significance for the science of water management. Because the understanding of source/transport/fate (i.e. pathways) of sediment-associated nutrients and contaminants is relevant to diffuse and point source management, the design of surveillance programmes must be substantially altered to capture the geochemical information represented in suspended sediment.

SAMPLING STRATEGIES

Bed vs. suspended sediment for geochemical purposes

Fine-grained suspended sediment can be used to indicate nutrient and contaminant stress in aquatic ecosystems. Whereas lacustrine bottom sediments can be used to assess cumulative chemical stress, there is some doubt whether this is equally true for river bottom sediments. Förstner & Salomons (1980), in their review of sediment geochemistry, suggest that bottom sediments in fluvial systems are an appropriate medium for monitoring heavy metal stress. Data in Ongley *et al.* (1981) and in Table 1 suggest that Förstner & Salomons' view may not be entirely correct. For a variety of Canadian rivers in many different physical settings there is clearly little relationship even at high flow conditions noted here, between particle-size characteristics of suspended and bottom sediments. Those rivers with large proportions of bottom material <62 μm (e.g. Carrot, Assiniboine, Red) are situated on fine-grained glaciolacustrine sediments which should not be confused with contemporary alluvial deposits. Unlike many European rivers, North American rivers are not lock-controlled and often display relatively mobile beds, responding to storm and seasonal flow regimes. There is, therefore, some doubt whether bottom sediments either reflect the geochemistry of suspended matter in transit about the bed, or characterize cumulative (time integrated) stress over longer periods. It is for this reason that this author has resorted to using macro-algae and macrophytes to obtain time integrated data on nutrient and contaminant stress in fluvial systems (Bjerkelund & Ongley, 1980; Ongley & Blachford, 1982).

Sampling technique

As illustrated by Culbertson *et al.* (1972), Ongley *et al.* (1981) and Table 2, concentration of the <62 μm fraction of suspended sediment is not depth dependent. In practice this implies that samples extracted near the top of the water column are geochemically representative yet avoid undue collection of coarse-grained material which should, in any event, be removed prior to chemical analysis.

Representative bulk sampling of suspended matter is routinely performed in the field by a portable continuous-flow centrifuge system developed in the course of the author's research (Allan, 1979; Ongley, 1978, Ongley *et al.*, 1981). This particular system has been calibrated with respect to recovery characteristics (Ongley & Blachford, 1982) and is now in use by a variety of government agencies for routine monitoring and research purposes. Alternative

TABLE 1 Average particle size characteristics of suspended and bed materials for selected high flow events[1] *in some Canadian rivers, 1978 and 1979*

		---Depth-Integrated Samples---			-Bed Material-	
River	Yr	N[2]	%>2μm	%>62μm	N[3]	%>62μm
Fraser (B.C.)						
(08MH024)[4]	'79	3	9.3 (06-13)[5]	69.3 (68-71)	5	00
(08MF035)	'78	4	9.8 (05-17)	50.2 (30-66)	6	00
(08MC018)	'78	3	11.7 (09-13)	70.0 (63-75)	5	0.5
Assiniboine (Man)						
(05MJ001)	'79	15	29.8 (15-40)	93.6 (85-97)	5	05
(05MH005)	'79	9	16.8 (09-24)	71.4 (54-86)	5	02
(05MJ003)	'79	2	27.0 (21-33)	82.0 (70-94)	5	36
(05MJ003)	'78	3	29.0 (27-31)	89.0 (86-91)	5	00
Red (Man.)						
(05OC001)	'79	5	44.0 (35-66)	97.4 (96-98)	5	57
(05OC012)	'78	2	48.5 (35-62)	97.5 (97-98)	5	13
Turtle (Man.)						
(05LJ007)	'79	8	25.9 (22-31)	81.6 (70-93)	5	00
Whitemud (Man.)						
(05LL005)	'79	1	41	88	5	02
Oldman (Alta.)						
(05AD007)	'78	2	13.0 (12-14)	76.0 (67-85)	7	23
Carrot (Sask.)						
(05KC001)	'78	1	57	95	7	48
N. Sask. (Sask.)						
(05GG001)	'78	2	18.5 (17-20)	66.0 (66-66)	2	00
Ausable (Ont.)						
(02FF002)	'79	2	63.0 (58-68)	98.5 (97-100)	5	13
(02FF002)	'78	1	76	97	7	01
Big Otter (Ont.)						
(02GC026)	'78	3	8.0 (07-09)	45.7 (44-47)	7	08
Canagagigue (Ont.)						
(02GA036)	'79	1	34	97	5	46
(02GA036)	'78	-	-	-	5	25
April Brk. (N.S.)						
(01FB005)	'78	1	4.0	86	1	24
Annapolis (N.S.)						
(01DC005)	'78	-	-	-	1	05
Pembroke (N.S.)						
(01DG018)	'78	-	-	-	1	01
Kennebecasis (NB)						
(01AP004)	'78	-	-	-	1	02
N.Nashwaaksis(NB)						
(01AK005)	'78	-	-	-	1	06
Brudenell (PEI)						
(01CE003)	'78	-	-	-	7	04
Emerald Brk.(PEI)						
(01CB006)	'78	-	-	-	7	05
North Brk. (PEI)						
(01CB005)	'78	1	14	95	7	05
Wilmot (PEI)						
(01CB004)	'78	2	4 (02-06)	74.0 (52-96)	7	07

[1] Spring melt and other selected high-flow events.

[2] Number of days sampled.

[3] Number of bottom samples on one or more days.

[4] Inland Waters Directorate station location code.

[5] Range of values reported

Source: Water Survey of Canada, 1980; 1981.

TABLE 2 Concentration of <62 µm suspended sediment fraction at various depths for Fraser River at Mission (B.C.) Station[1] 08MH024, May 29 and June 06, 1979

Depth (m)[4]	*Concentration*		*62 mm (ppm)*	
	May 29[2]		*June 06*[3]	
0.15	*338*	*(88)*[5]	*188*	*(79)*[5]
2.38	*360*	*(76)*	*198*	*(68)*
4.75	*367*	*(73)*	*197*	*(62)*
7.16	*367*	*(64)*	*185*	*(50)*
9.54	*378*	*(54)*	*209*	*(36)*
10.73	*358*	*(42)*	*211*	*(35)*
11.31	*370*	*(33)*	*210*	*(34)*
11.76 (stream bed)		*(0)*[6]		*(0)*[6]

[1] *Data from Water Survey of Canada (1981).*

[2] *Daily mean discharge May 29 = 6880* $m^3 s^{-1}$ *exceeded by only 3.6% of daily flows in 1979.*

[3] *Daily mean discharge June 6 = 7620* $m^3 s^{-1}$ *exceeded by only 1.0% of daily flows in 1979.*

[4] *Section is mid-channel (274.3 m location).*

[5] *<62 µm fraction as per cent of total measured suspended sediment*

[6] *<62 µm fraction as per cent of bottom sediment (mean of five samples).*

bulk collection procedures include laboratory centrifugation of bulk samples, filtration, flocculation and settling. These latter methods tend to have moderate to severe physical or chemical limitations for routine field collection of suspended matter.

Hysteresis effects upon sediment geochemistry

The hysteresis effect between suspended sediment concentration and the runoff hydrograph is now well known (Walling, 1977). As a consequence, sediment sampling is usually focussed on the rising limb of the hydrograph where concentrations tend to be significantly higher than at comparable discharge on the falling limb. Much less is known, however, about the geochemical variation during a runoff event (Turner *et al.*, 1977). Limited data from Bynoe (1979) suggest that for one small nonpoint source watershed (Wilton Creek, noted below) in southeastern Ontario, there is increased concentration of Mn, Cd, Zn, Ni and Cu on the falling limb even though sediment concentration is reduced (Fig.2). Unexpectedly, the silt/clay ratio increases considerably on the falling limb, implying a coarsening of suspended matter. This is, however, accompanied by an increase in organic content. These observations suggest that conventional sediment sampling strategies need to be re-examined for chemical purposes.

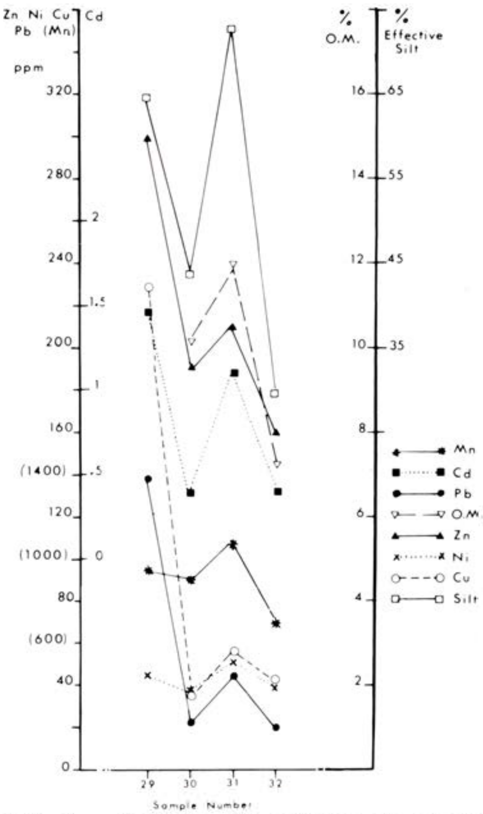


FIG.2 Hysteresis effect in suspended sediment quality. Samples 30 and 31 are rising and falling limb respectively. Samples 29 and 32 are sediment concentrations respectively for preceding and following storm flows (after Bynoe, 1979)

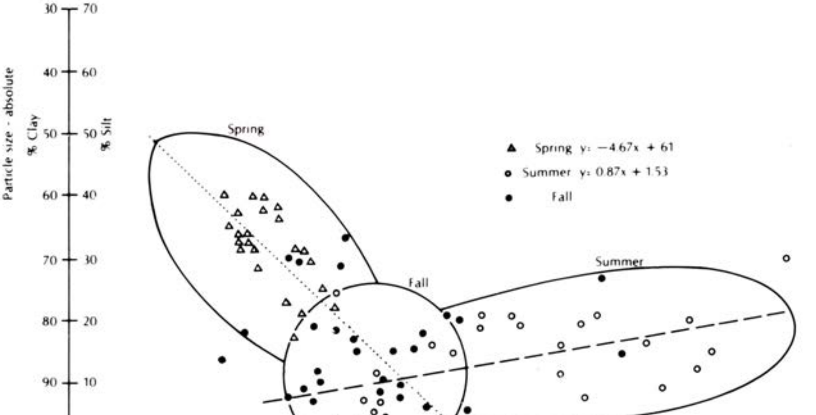
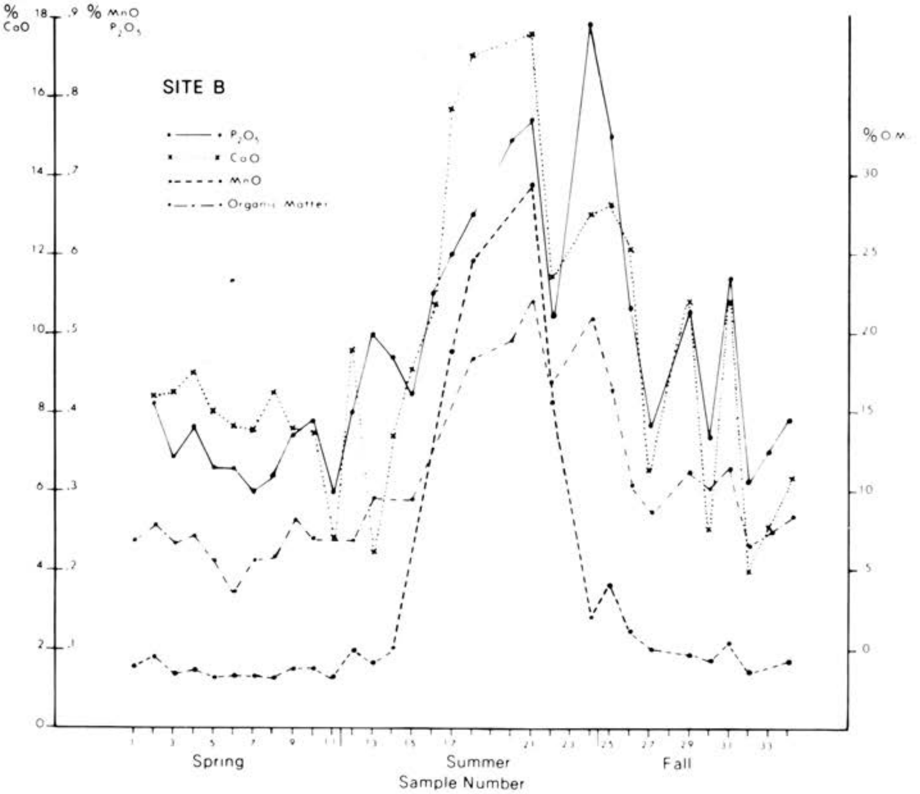


FIG.3 Seasonal effects on suspended sediment size and organic content (after Ongley et al. 1981)



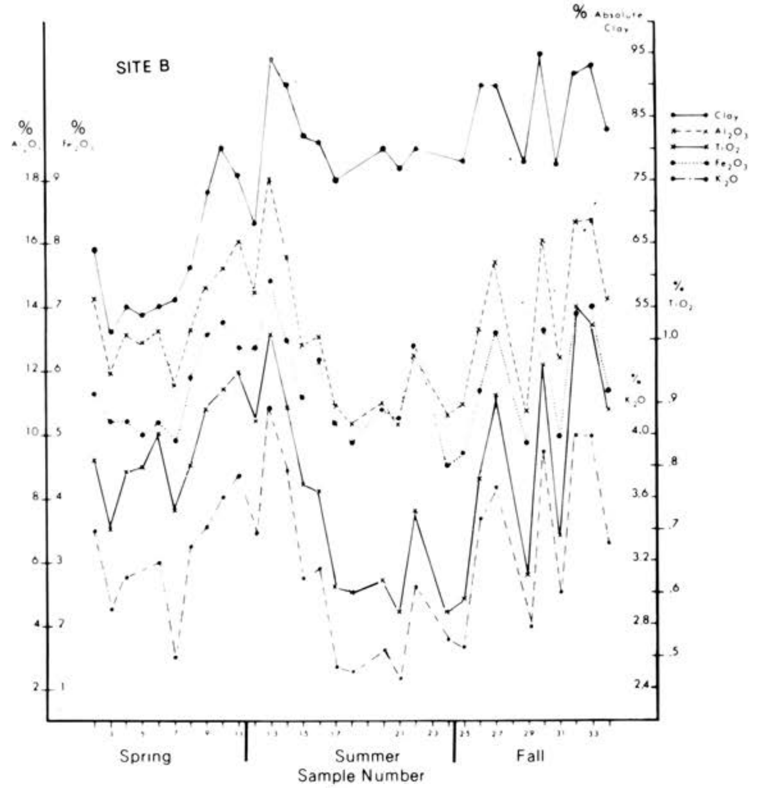


FIG.4 Seasonal distribution of sediment-associated elements (after Ongley et al. 1981)

INFLUENCE OF SOURCE AND SEASONALITY OF SUSPENDED SEDIMENT

Mineralogy of suspended sediment has been used with varying degrees of success to identify sediment source areas (Packham *et al.*, 1960; Moore, 1961; Lund *et al.*, 1972). Whereas Wall & Wilding (1976) were able to differentiate between stream bank and surficial erosion on the basis of presence/absence of carbonates during low/high flow events on the Maumee River, Angino & Schneider (1975) found mineralogy to be essentially similar throughout the flow regime of selected Kansas streams. In our work on storm flow on Wilton Creek, a 132 km^2 representative drainage basin in southeastern Ontario (Percival, 1979; Ongley *et al.*, 1981) we found little variation in mineralogy over a complete year.

In contrast, we found that concentrations of major elements (as oxides) on suspended sediments and the shift in organic/mineral ratio appear to be closely related to sediment source. All samples noted in Figs 3 and 4 were obtained during spring melt and most stormflow events for one year. Samples were collected with the continuous-flow centrifugation equipment noted above. Organic matter and silt/clay ratio are clearly seasonally dependent (Fig.3) and indicate that coarser sediment (also verified by sand content) is transported during periods of expanded contributing area in spring. Organic matter (plankton, decay products of *Cladophora* sp. and macrophytes) becomes significant during the summer period when there is extensive bioproductivity within the stream.

Analysis of major elements by X-ray fluorescence indicates that peaks of P, Ca and Mn (Fig.4) coincide with the major period of biomass production. Concentrations of these three elements in representative soil and bed/band samples are significantly lower, indicating accumulation of these elements in stream biomass with subsequent release into the suspended load in the form of organic detritus and decay products.

In contrast with Ca, P and Mn, oxides of Al, Fe, Ti and K (Fig.4) in spring samples compare closely with levels found in soils. This suggests that erosion of soil surfaces during spring is the major contributor of suspended matter. Concentrations of these elements decline in summer when suspended sediment is more influenced by in-stream organic detritus. Autumn data are variable, indicating pronounced fluctuation in runoff intensity for autumn storm events.

DISTANCE EFFECTS ON SUSPENDED SEDIMENT QUALITY

It is now well established that there are significant losses of, for example, phosphorus as one moves downstream from a major point source. Although the mechanisms are thought to be adsorption, coprecipitation and bioaccumulation, the rate of depletion (distance decay effect) of elements from water and the consequent changes in sediment and biomass chemistry are difficult to quantify. Verhoff *et al.* (1978) use kinematic wave theory to explain observed distance effects upon total phosphorus transport during storm events for a 270 km distance on the Sandusky River, Ohio.

In our study of Wilton Creek, event sampling at four sites located some 1500-2000 m apart failed to detect between-site differences in sediment chemistry. This suggests that downstream geochemical change should occur over much longer distances. Preliminary results from long-distance pathway analysis over a 1 year period for total Zn, Hg and P from suspended sediment and Zn in biomass are presented in Fig.5 for the Bow River, Alberta, for summer 1980.

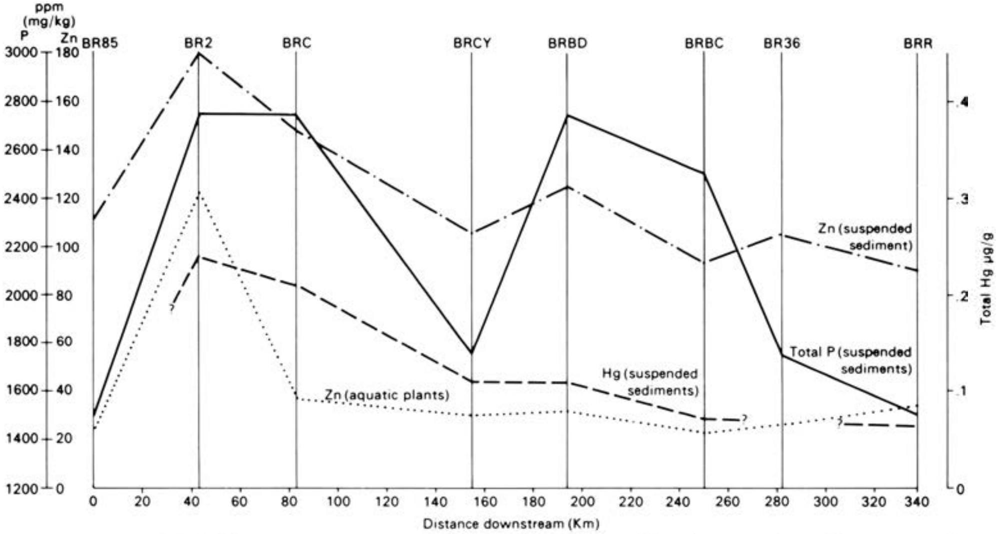


FIG.5 Downstream distribution of zinc, phosphorus and mercury associated with suspended sediment and rooted plants, Bow River, Alberta.

Samples characterize seasonal discharge regimes and were collected sequentially downstream to reflect time of travel. Site BR85 represents essentially background as the Bow River enters the Calgary metropolitan area. There are two major sewage treatment plants discharging into the Bow River between BR85 and BR2. There are no point sources downstream from BR2 to the confluence of the Bow and Oldman Rivers below BRR, 340 km downstream from BR85. The only complicating factor is the dam and silted-up reservoir at Bassano (BRBD). In general, one can see a sudden increase in all elements immediately downstream from Calgary, then a progressive decrease. All elements, whether on suspended sediment or associated with biomass, display the same relative change. The increase at BRBD may be related to phytoplankton growth in the reservoir.

Until other physical and chemical data are complete one cannot offer an explanation for the downstream reduction of elements associated with the wash load. One can speculate that downstream reduction may be influenced by input on uncontaminated sediment to the river with consequent dilution of sediment-associated concentrations of phosphorus and metals. It is significant that bioaccumulation of Zn by (predominantly) macrophytes mirrors changes noted in the suspended sediment fraction. Apart from filtering out Zn from the water column, biomass data suggest that patterns of bioconcentration may be useful in characterizing the spatial component of nutrient and contaminant stress in the fluvial environment.

ACKNOWLEDGEMENTS Material in this paper is drawn from a variety of research programmes variously funded by the Natural Sciences and Engineering Research Council of Canada. Inland Waters Directorate of Environment Canada and the Toxic Contaminants Management Programme (Environment Canada) while the author was at Queen's University. The assistance and collaboration of present and past graduate students are gratefully acknowledged.

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