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Annual suspended sediment and trace element fluxes in the Mississippi, Columbia, Colorado, and Rio Grande drainage basins

Arthur J. Horowitz,* Kent A. Elrick and James J. Smith
US Geological Survey, Peachtree Business Center, Suite 130, 3039 Amwiler Road, Atlanta, GA 30360, USA

Abstract:

Suspended sediment, sediment-associated, total trace element, phosphorus (P), and total organic carbon (TOC) fluxes were determined for the Mississippi, Columbia, Rio Grande, and Colorado Basins for the study period (the 1996, 1997, and 1998 water years) as part of the US Geological Survey's redesigned National Stream Quality Accounting Network (NASQAN) programme. The majority ($\geq$70%) of Cu, Zn, Cr, Ni, Ba, P, As, Fe, Mn, and Al are transported in association with suspended sediment; Sr transport seems dominated by the dissolved phase, whereas the transport of Li and TOC seems to be divided equally between both phases. Average dissolved trace element levels are markedly lower than reported during the original NASQAN programme; this seems due to the use of 'clean' sampling, processing, and analytical techniques rather than to improvements in water quality. Partitioning between sediment and water for Ag, Pb, Cd, Cr, Co, V, Be, As, Sb, Hg, and Ti could not be estimated due to a lack of detectable dissolved concentrations in most samples.

Elevated suspended sediment-associated Zn levels were detected in the Ohio River Basin and elevated Hg levels were detected in the Tennessee River, the former may affect the mainstem Mississippi River, whereas the latter probably do not. Sediment-associated concentrations of Ag, Cu, Pb, Zn, Cd, Cr, Co, Ba, Mo, Sb, Hg, and Fe are markedly elevated in the upper Columbia Basin, and appear to be detectable (Zn, Cd) as far downstream as the middle of the basin. These elevated concentrations seem to result from mining and/or mining-related activities. Consistently detectable concentrations of dissolved Se were found only in the Colorado River Basin.

Calculated average annual suspended sediment fluxes at the mouths of the Mississippi and Rio Grande Basins were below, whereas those for the Columbia and Colorado Basins were above previously published annual values. Downstream suspended sediment-associated and total trace element fluxes increase in the Mississippi and Columbia Basins, whereas fluxes markedly decrease in the Colorado Basin. No consistent pattern in trace element fluxes was detected in the Rio Grande Basin.

KEY WORDS sediment chemistry; flux; trace elements; suspended sediment

INTRODUCTION

In 1994, in response to substantially diminishing resources, to changes in data requirements, and to better integrate with other US Geological Survey (USGS) ambient water-quality monitoring programmes, the original National Stream Quality Accounting Network (NASQAN) programme was redesigned as a flux-based water-quality monitoring network for the Mississippi, Columbia, Colorado, and Rio Grande drainage basins (Hooper *et al*., 2001). There are two questions/issues related to trace elements that are common to all four basins. (1) What are the annual trace element fluxes within each NASQAN basin (or subbasin where appropriate) and to the coastal zone, and how are they partitioned between the dissolved and solid phase? (2) Are there any dissolved or solid phase-associated trace element concentrations in the NASQAN basins that may represent a potential problem for drinking water supplies, recreation, or aquatic life? The answers to

* Correspondence to: A. J. Horowitz, US Geological Survey, Peachtree Business Center, Suite 130, 3039 Amwiler Road, Atlanta, GA 30360, USA. E-mail: horowitz@usgs.gov

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these two questions to date, based on the first 3 years of the NASQAN programme, are provided herein for each basin. The procedures/protocols used to determine dissolved and suspended sediment-associated trace element concentrations, as well as those employed to estimate annual fluxes and their associated errors are described elsewhere (Horowitz *et al.*, 2001). Readers should note that as a result of a lack of analyses (Hg, Ti) and/or analytical sensitivity (Ag, Pb, Cd, V, Se, Sb, Mo), total trace element fluxes (dissolved plus suspended sediment-associated) could not be estimated for several constituents.

Each of the four NASQAN basin sampling designs was established with a view toward understanding and evaluating the movement of water and a variety of water-quality-associated parameters. Schematic diagrams of the sampling locations are provided for each basin in Figures 1–4. In general, and particularly in large rivers where individual point sources (e.g. an industrial outfall) only tend to have relatively local impacts, water and dissolved trace elements tend to behave conservatively; therefore, large river segments tend to be fairly compositionally homogeneous. Thus, contributions from sizeable portions of a basin, and/or potential sources or sinks for a variety of chemical parameters, can be estimated by adding/subtracting the contributions/losses of large river segments. For example, fluxes for the Missouri River can be determined by direct measurement at Hermann, whereas the impact of these contributions to Mississippi River fluxes can be determined by subtracting the fluxes determined for Clinton from those determined at Thebes (Figure 1). Or, in other words, the fluxes at Thebes should be the sum of those at Hermann and Clinton. Further, the injection of a substantial quantity of a hydrophilic substance will tend to move through a large river basin along with the water itself. Hence, continued sampling of the same segment of water (i.e. Lagrangian sampling) as it moves through the system will permit the delineation of ongoing concentration changes and potentially, the physical/chemical processes acting on that substance to engender those compositional changes. Finally, the travel time for the impacted 'segment' can be readily determined/predicted if the discharge or water velocity is known.

On the other hand, suspended sediment does not behave conservatively; it moves in and out of suspension, and there are constant exchanges between the water column, the river bed, and the river banks. Thus, the particles making up a 'packet' of sediment, and their associated chemical constituents, continuously change as material moves in (deposition) and out (resuspension) of 'storage' while the packet is transported downstream. As such, the packet rarely retains its original composition, even over relatively short distances, and travel times are very difficult to predict. Thus, suspended sediment and sediment-associated chemical constituents display much more marked spatial and temporal variability than dissolved constituents (e.g. Horowitz, 1995). Hence, continued sampling of the same packet of water as it moves downstream will not readily permit the delineation of ongoing particle-specific concentration changes, nor potential physical/chemical processes

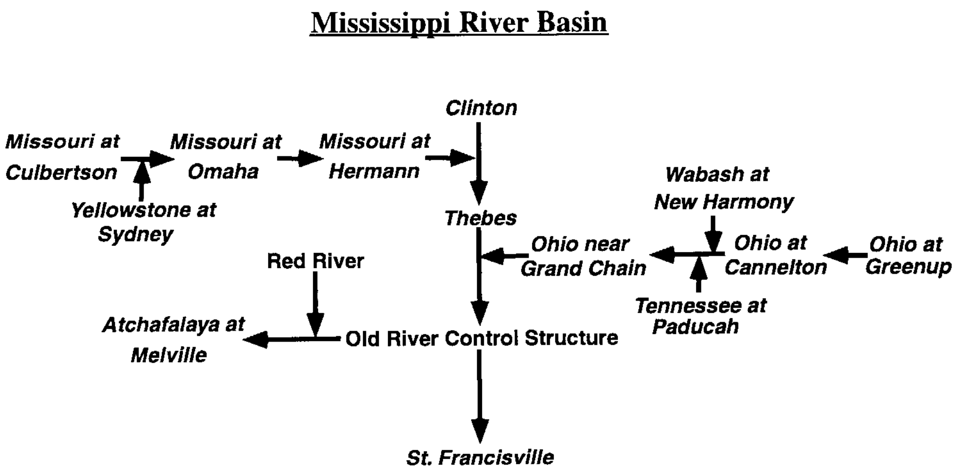

Figure 1. A schematic diagram of the NASQAN Mississippi River Basin; italicized sites represent actual sampling sites (not to scale). The arrows indicate downstream flow direction

acting on that sediment-associated substance. As a result of this nonconservative behaviour, contributions from sizeable portions of a basin, and/or potential sources or sinks for a variety of sediment-associated chemical parameters, cannot be reasonably estimated by adding/subtracting the contributions/losses of large river segments.

As a result of the differences in the physical/chemical behaviour of water and suspended sediment, a sampling programme designed to monitor dissolved fluxes may not be applicable to monitoring sediment and sediment-associated chemical fluxes. This can and does create major network design problems, particularly when funding is limited. As an example of the impact of these differences, examine the sampling scheme for the middle/lower Mississippi River [from Thebes to St. Francisville, including the location of the Old River Control Structure, as well as the location of the Melville sampling site on the Atchafalaya River (Figure 1)]. Note that there are no mainstem sampling sites below the confluence of the Ohio and the Mississippi Rivers before St. Francisville, a distance of over 1100 km. Between the Ohio and Mississippi confluence and St. Francisville, the Arkansas River joins the Mississippi River, and about 25% of the Mississippi River is diverted into the Atchafalaya River at the Old River Control Structure (Figure 1). Further, the Atchafalaya sampling site (Melville) is downstream from its confluence with the Red River. As such, the effects of suspended sediment and the sediment-associated chemical contributions from the Ohio and the Arkansas Rivers on the mainstem Mississippi River cannot be readily delineated. Further, due to the location of the Melville sampling site and the lack of a sampling site at or near the Control Structure, there is no way to separate out the suspended sediment and suspended sediment-associated trace element contributions to the Atchafalaya between the Mississippi and Red Rivers. Water chemists resolve this problem by assuming that dissolved constituents behave conservatively; thus, the dissolved concentrations determined at St. Francisville are assumed to be equivalent to those in Mississippi River water diverted by the Control Structure. On the other hand, sediment chemists cannot make this assumption. In fact, substantial sediment resuspension has been observed between the Control Structure and the St. Francisville sampling site (R. H. Meade, USGS, oral communication, 1999). Readers should bear these various considerations/caveats/limitations in mind when examining the results for suspended sediment and sediment-associated trace element fluxes in the four NASQAN basins reported and discussed herein.

The annual sediment and dissolved chemical fluxes estimated for this study were calculated by summing a series of daily instantaneous fluxes for each water year (October 1 through September 30) using the following formula:

$$\text{flux (tonnes day}^{-1}) = [\text{Q (m}^3\ \text{s}^{-1})][\text{conc. (mg l}^{-1})][0{\cdot}0864] \tag{1}$$

where Q = discharge, conc. = suspended sediment concentration, dissolved constituent concentration.

The suspended sediment-associated trace element, phosphorus (P) and carbon (C) fluxes were estimated using the same formula after recalculation of the concentration from mass mass^{-1} units to mass volume^{-1} units as follows:

$$\text{TE conc. (mg l}^{-1}) = [\text{TE conc.}(\mu\text{g g}^{-1})][\text{TSS conc. (g l}^{-1})]/1000 \tag{2}$$

where TE conc. = trace element concentration, TSS conc. = total suspended sediment concentration.

The errors associated with the discharge measurements (Q) typically are $\pm 3-5\%$ (Sauer and Meyer, 1992). Actual suspended sediment concentration measurements, based on replicate samples, as well as interlaboratory measurements of standards, normally are less than $\pm 10\%$; however, the vast majority of suspended sediment concentrations used in the calculations are model-derived, and have varying degrees of error associated with them depending on the site as well as the discharge (Horowitz *et al.*, 2001). Finally, the chemical concentrations, based on the simultaneous analysis of standard reference samples normally fall within $\pm 10\%$; however, the chemical concentrations used in the flux calculations were derived mean/median concentrations on a site-by-site basis and display errors ranging from $\pm <1\%$ to 75% (Horowitz *et al.*, 2001). Although the appropriate number of significant figures were used in the individual instantaneous daily flux calculations, there was no way to adequately assess the errors associated with either the individual or the final summed flux results; as such, the actual final sums are provided in the tables.

THE MISSISSIPPI RIVER BASIN

Basic geographic data covering the Mississippi River and its major tributaries are provided in Table I, and a schematic of the sampling sites is provided in Figure 1. Mean and median chemical concentrations for suspended sediment-associated trace elements from the basin are provided in Table II. With few exceptions, suspended sediment-associated trace element concentrations for the mainstem Mississippi River and its tributaries are typical for fine-grained sediment (Table II) (Horowitz, 1991; Persaud *et al*., 1993). However, Zn seems to be substantially elevated (by about a factor of two) in the Ohio River and some of its tributaries relative to typical background levels. Zn levels are elevated at Grand Chain, hence, it is likely that Zn-rich suspended sediment from the Ohio River Basin reaches the Mississippi River. These elevated levels may be associated either with water from the Allegheny and/or Monongahela Rivers, which drain the coalfields in Pennsylvania prior to joining the Ohio River, and/or from heavy industry in the Ohio River Basin. In addition, suspended sediment from the Tennessee River seems enriched in Hg (Table II) by about a factor of two relative to typical background levels (Horowitz, 1991; Persaud *et al*., 1993). However, Hg levels are not elevated at Grand Chain; hence, it is unlikely that Hg-rich suspended sediment from the Tennessee reaches the Mississippi River. The elevated Hg levels may have resulted from past discharges from the Y-12 Facility at Oak Ridge, TN (Carmichael, 1989).

Annual suspended sediment fluxes to the Gulf of Mexico (Mississippi River at St. Francisville plus Atchafalaya at Melville) for each year of the study period averaged about 190 Mt. This seems somewhat low (−17%) compared to previously published averages, but within the range of natural variability (Tables I and III).

Table I. General geographic information about the rivers and major tributaries in the four NASQAN basins

River	Length[a] (km)	Drainage area (10^3 km^2)	Discharge[a] (10^3 m^3 s^{-1})	Suspended sediment ($\times 10^6$ t year^{-1})[b]
Mississippi River	3782	2979	16·8	210–230[c,d]
Missouri River	4076	150	2·16	95[d]
Ohio River	2101	526	7·96	40[d]
Wabash River	798	74·1	0·75	7[d]
Tennessee River	1448	106	1·93	2[d]
Atchafalaya[g]	2285	246	1·64	80[d,e]
Columbia River	1996	668	7·5	10[c]
Snake River	1670	280	1·61	2[f]
Willamette River	497	29·5	1·06	1·1[f]
Rio Grande	3033	870	0·03	0·7 ± ·5[e] (1967–1983)
Colorado River	2334	637	0·41	0·1[c]
Green River	1175	105	0·19	8·8 ± 4·4[d] (1965–1984)
San Juan River	640	31	0·07	15·4 ± 7·8[d] (1953–1967)

[a] van der Leeden *et al*. (1990).
[b] Megatonnes.
[c] Meade and Parker (1985).
[d] Meade (1995).
[e] USGS Suspended Sediment Database (1999).
[f] Haushild *et al*. (1966).
[g] Including the Red River, but excluding Mississippi River water diverted at the Old River Control Structure.

Table II. Mean (Mn)/median (Md) trace element concentrations for suspended sediment collected at all the NASQAN sampling sites

Sample location		n	mg l^{-1}																				wt%				
			Ag	Cu	Pb	Zn	Cd	Cr	Co	Ni	Ba	V	Li	Be	Mo	P	Sr	As	Sb	Se	Hg	Mn	Fe	Al	Ti	TOC	Total C
Background[a]				25	23	65	1·1	31		31						<600		4·2			0·10	400	3·1			<1·0	
Background[b]			<1·0	20	23	90	<1·0	50	20	25								7	1·2	1	0·10	600	2·8	5·5	0·41	1·4	
Ohio River at	Mn	23	<0·5	42	44	260	0·8	95	32	81	520	99	64	2·7	<5	1200	210	15·5	1·6	1·3	0·09	2600	4·4	7·5	0·42	3·6	3·9
Greenup Dam	Md		<0·5	40	43	240	0·8	92	33	79	540	100	66	2·9	<5	1200	170	14·6	1·3	1·1	0·10	2600	4·6	7·9	0·44	3·6	3·9
Ohio River at	Mn	36	<0·5	41	38	200	0·6	91	25	67	500	100	60	2·4	<5	1400	190	14·0	2·2	1·0	0·08	1900	4·1	7·4	0·44	2·8	3·2
Cannelton Dam	Md		<0·5	36	37	190	0·6	80	24	59	520	110	59	2·5	<5	1400	140	14·0	1·4	0·9	0·08	1800	4·2	7·8	0·47	2·8	3·2
Wabash River at	Mn	26	<0·5	25	26	140	0·7	64	13	39	480	91	42	1·6	<5	1200	150	11·0	1·5	0·9	0·14	1400	3·3	6·2	0·37	2·5	3·6
New Harmony	Md		<0·5	25	25	140	0·5	65	13	40	480	92	39	1·5	<5	1200	130	11·0	1·0	0·8	0·06	1200	3·3	6·3	0·38	2·4	3·2
Tennessee River	Mn	24	<0·5	34	34	220	0·7	110	16	70	510	86	49	1·8	<5	1900	150	13·0	2·0	0·9	0·19	3300	3·5	6·4	0·38		4·2
at Paducah	Md		<0·5	29	33	220	0·7	96	17	63	490	86	45	1·8	<5	2000	140	12·0	1·8	0·8	0·18	3000	3·6	6·3	0·39		4·2
Ohio River	Mn	39	<0·5	34	36	190	0·7	79	21	56	510	100	56	2·2	<5	1400	130	14·0	1·8	0·8	0·11	2000	3·9	7·1	0·45	2·6	3·0
near Grand Chain	Md		<0·5	32	33	180	0·7	75	20	52	500	100	53	2·2	<5	1400	120	13·0	1·2	0·7	0·08	1800	4·0	7·1	0·45	2·6	2·9
Mississippi River	Mn	37	<0·5	21	32	140	0·6	74	12	37	550	80	29	1·3	<5	1500	150	8·0	2·4	1·0	0·17	2200	3·1	5·3	0·31	3·8	4·4
at Clinton	Md		<0·5	20	31	140	0·6	63	12	35	560	77	29	1·2	<5	1400	140	8·0	1·1	1·0	0·08	2200	3·0	5·2	0·31	3·6	4·4
Missouri River	Mn	16	<0·5	17	14	78	0·2	66	10	39	670	87	34	1·3	<5	600	270	14·0	2·4	0·4	0·03	360	2·4	5·5	0·24	0·8	1·5
near Culbertson	Md		<0·5	16	15	73	0·2	69	10	38	740	80	34	1·3	<5	610	270	14·0	2·1	0·4	0·03	340	2·3	5·3	0·24	0·8	1·5
Yellowstone River	Mn	26	<0·5	21	19	86	0·3	75	11	39	660	98	35	1·5	<5	730	270	12·0	2·1	0·6	0·04	540	2·8	6·4	0·29	1·2	1·8
near Sidney	Md		<0·5	21	18	81	0·3	70	11	36	660	93	34	1·4	<5	740	260	12·0	1·2	0·6	0·04	520	2·8	6·2	0·28	1·1	1·7
Missouri River at	Mn	43	<0·5	13	16	60	0·4	53	9	36	630	60	23	1·0	<5	650	240	9·0	1·8	0·8	0·03	720	1·8	4·6	0·19	1·1	1·5
Omaha	Md		<0·5	11	16	54	0·4	53	8	35	700	51	22	0·9	<5	610	250	8·0	1·2	0·7	0·03	700	1·7	4·3	0·17	1·0	1·5
Missouri River at	Mn	41	<0·5	18	21	77	0·5	56	10	34	660	77	29	1·3	<5	750	210	9·0	1·4	1·0	0·04	730	2·3	5·4	0·25	1·7	1·9
Hermann	Md		<0·5	17	20	74	0·4	56	9	33	680	80	29	1·3	<5	760	210	9·0	1·1	0·8	0·03	760	2·3	5·2	0·25	1·5	1·8
Mississippi River	Mn	43	<0·5	21	26	93	0·5	67	10	41	610	80	31	1·3	<5	910	200	9·0	2·4	1·0	0·04	910	2·5	5·5	0·29	2·2	2·6
at Thebes	Md		<0·5	21	24	90	0·5	63	11	38	610	82	31	1·3	<5	890	180	9·0	1·2	0·9	0·04	950	2·5	5·5	0·30	1·9	2·4
Mississippi River	Mn	42	<0·5	23	26	110	0·5	65	13	40	600	86	35	1·6	<5	940	160	11·0	1·7	0·6	0·05	1200	2·9	6·1	0·33	1·6	1·9
at St. Francisville	Md		<0·5	22	24	110	0·5	64	13	39	600	88	36	1·6	<5	980	150	11·0	1·1	0·6	0·04	1100	3·0	6·1	0·33	1·5	1·8
Atchafalaya River	Mn	40	<0·5	20	23	100	0·5	65	12	39	560	85	36	1·6	<5	910	150	10·0	2·4	0·6	0·05	1200	2·8	5·9	0·32	1·5	1·8
at Melville	Md		<0·5	21	22	100	0·4	61	12	37	570	85	36	1·6	<5	900	140	11·0	0·9	0·5	0·05	1000	2·7	5·9	0·33	1·4	1·6
Columbia River at	Mn	11	4·2	880	230	5300	3·6	160	42	62	1200	76	31	1·7	24	1300	390	11·5	69	1·1	0·22	2000	7·7	5·5	0·23	2·6	3·0
Northport	Md		4·5	680	220	6000	3·4	140	42	48	1200	77	31	1·7	22	1300	420	12·0	69	1·2	0·19	2100	7·1	5·4	0·24	2·0	3·4

(*continued overleaf*)

Table II. (*continued*)

Sample location		n	mg l^{-1}																			wt%				
			Ag	Cu	Pb	Zn	Cd	Cr	Co	Ni	Ba	V	Li	Be	Mo	P	Sr	As	Sb	Se	Hg	Mn	Fe	Al	Ti	TOC	Total C
Columbia River at	Mn	5	<0·5	74	110	570	2·9	160	19	120	700	100	32	1·6	<5	2000	320	16·7	3·0	1·1		1900	4·1	6·1	0·37	4·1	5·5
Vernita Bridge	Md		<0·5	68	89	500	2·7	160	20	120	660	97	30	1·4	<5	1600	280	14·8	2·3	1·1		1800	4·1	5·9	0·35	4·1	5·4
Snake River at	Mn	22	<0·5	49	35	160	0·5	63	22	39	650	120	39	1·9		1500	230	11·0	1·8	0·6	0·08	1500	5·0	7·5	0·54	3·3	3·5
Burbank	Md		<0·5	43	24	130	0·4	54	22	35	650	120	37	1·9		1400	220	11·0	1·3	0·6	0·08	1400	5·0	7·5	0·54	3·2	3·2
Columbia River at	Mn	20	<0·5	53	28	220	1·3	66	19	42	610	120	30	1·6	<5	1300	270	9·0	1·6	0·5	0·11	1200	4·7	7·1	0·51	2·6	3·4
Warrendale	Md		<0·5	49	29	220	1·3	62	18	40	610	120	30	1·6	<5	1300	270	9·0	1·5	0·5	0·07	1200	4·6	7·2	0·50	2·8	3·3
Willamette River	Mn	40	<0·5	64	27	140	0·3	85	26	51	460	160	29	1·4	<5	1900	240	8·0	1·0	0·4	0·09	2100	5·8	8·2	0·68	3·5	4·0
at Portland	Md		<0·5	56	23	140	0·3	79	25	46	460	160	26	1·5	<5	1800	230	8·0	1·0	0·4	0·08	1800	5·9	8·3	0·68	3·6	4·0
Columbia River	Mn	39	<0·5	50	25	160	0·8	59	18	38	560	110	28	1·5	<5	1300	320	7·0	1·1	0·4	0·07	1100	4·4	7·8	0·51	2·2	2·5
near Beaver Army Terminal	Md		<0·5	49	23	170	0·8	57	18	36	560	110	27	1·5	<5	1200	310	7·0	1·0	0·3	0·07	1100	4·3	7·8	0·50	2·0	2·1
Rio Grande at	Mn	25	<0·5	19	20	85	0·3	43	7	25	500	42	47	1·3	<5	700	490	5·0	2·4	0·3	0·03	1200	1·8	4·8	0·23	1·4	2·8
El Paso	Md		<0·5	20	17	79	0·3	43	8	24	580	45	45	1·4	<5	760	470	5·0	0·9	0·3	0·03	1000	1·9	5·3	0·25	1·1	2·8
Rio Grande at	Mn	31	<0·5	15	17	85	0·3	45	8	25	290	84	43	1·6	9	730	740	8·0	1·9	0·8	0·14	490	2·4	5·8	0·27	1·3	4·1
Foster Ranch	Md		<0·5	15	17	89	0·4	41	8	21	290	86	41	1·6	7	790	770	8·0	1·0	0·7	0·05	460	2·4	6·1	0·28	1·1	4·1
Rio Grande at	Mn	10	<0·5	14	18	110	0·4	37	7	17	380	72	36	1·2	<5	950	570	8·0	1·2	0·6	0·06	460	2·2	5·3	0·30	1·8	4·8
Laredo	Md		<0·5	14	18	100	0·3	36	7	17	360	64	36	1·1	<5	890	580	6·5	1·1	0·7	0·06	440	2·1	5·2	0·27	1·7	4·7
Arroyo Colorado	Mn	30	<0·5	13	16	76	0·5	26	5	16	210	43	41	0·9	<5	750	900	5·0	2·2	0·5	0·10	690	1·5	3·5	0·16	1·4	3·0
at Harlingen	Md		<0·5	11	12	66	0·3	24	5	16	200	37	42	0·9	<5	700	950	5·0	1·6	0·5	0·06	690	1·4	3·3	0·15	1·3	3·0
Rio Grande near	Mn	25	<0·5	16	26	110	0·4	35	7	22	370	64	44	1·3	<5	1200	720	11·0	2·9	0·8	0·14	1600	2·3	4·8	0·22	2·8	5·1
Brownsville	Md		<0·5	15	23	110	0·4	35	7	22	400	65	45	1·4	<5	1200	660	11·0	2·1	0·7	0·11	1500	2·3	5·0	0·23	2·8	5·0
Colorado River	Mn	30	<0·5	19	26	96	0·7	58	7	33	410	68	35	1·1	<5	610	490	7·0	3·1	1·9	0·04	440	1·8	4·5	0·20	1·6	3·2
near Cisco	Md		<0·5	18	24	95	0·7	53	7	28	400	70	34	1·2	<5	600	400	6·0	1·6	1·7	0·04	460	1·9	4·6	0·21	1·5	3·0
Green River at	Mn	32	<0·5	17	17	71	0·4	59	7	32	510	63	38	1·3	<5	670	460	7·0	2·7	0·6	0·03	350	1·9	4·9	0·21	1·0	2·4
Green River	Md			16	16	68	0·4	55	7	29	500	56	36	1·2	<5	680	300	6·0	0·9	0·5	0·02	350	1·8	4·6	0·20	1·0	2·2
San Juan River	Mn	23	<0·5	17	24	79	0·3	46	7	28	500	50	27	1·3	<5	390	370	5·0	1·6	0·4	0·03	410	1·9	5·3	0·22	0·8	1·5
near Bluff	Md			17	21	75	0·2	44	6	23	480	45	25	1·2	<5	400	280	5·0	0·9	0·3	0·02	400	1·6	4·8	0·21	0·8	1·3
Colorado River	Mn	15	<0·5	13	17	41	0·2	41	6	22	460	40	23	1·0	<5	400	170	4·7	1·1	0·2	0·02	330	1·6	4·4	0·20	0·5	1·8
above Diamond Creek	Md		<0·5	11	14	36	0·2	40	5	20	450	34	20	0·8	<5	420	160	4·5	0·9	0·2	0·01	310	1·2	3·7	0·18	0·5	1·8
Colorado River	Mn	13	<0·5	15	16	56	0·2	58	7	35	460	49	30	1·0	<5	640	460	6·0	4·5	0·8	0·04	1100	1·9	4·0	0·27	1·0	2·1
at NIB	Md		<0·5	15	14	52	0·2	59	8	34	460	49	28	0·9	<5	600	470	7·0	1·5	0·7	0·04	900	1·9	3·9	0·27	0·9	1·8

[a] Persaud *et al*. (1993).
[b] Horowitz (1991).

Table III. Summary of estimated Mississippi River Basin fluxes for the 1996, 1997, and 1998 water years

Location		S.S. (Mt)	Ag (t)	Cu (t)	Pb (t)	Zn (t)	Cd (t)	Cr (t)	Co (t)	Ni (t)	Ba (t)	V (t)	Li (t)	Be (t)	Mo (t)	P (t)	Sr (t)	As (t)	Sb (t)	Se (t)	Hg (t)	Fe (t)	Mn (t)	Al (t)	Ti (t)	TOC (t)	Total C (t)
Ohio River at Greenup	Flux (S)[1] WY:96	28·2	<14	1130	1210	6780	23	2600	900	2230	15 200	2820	1810	82	<141	33 900	4800	440	37	31	3	1 300 000	73 400	2 200 000	120 000	1 000 000	1 100 000
	Flux (D)[2] WY:96			140		290				190	3690		550		140	1680	14 800					4320	1840	1800		250 000	
	Flux (T)[3] WY:96			1270		7060				2420	18 900		2360			35 600	19 600					1 300 000	75 300	2 200 000		1 200 800	
	Solid (%)[4] WY:96			89		96				92	81		77			95	24					100	97	100		80	
	Flux (S) WY:97	14·4	<7	580	620	3450	12	1320	460	1140	7760	1440	920	42	<72	17 300	2440	220	19	16	1	660 000	37 400	1 100 000	63 300	520 000	560 000
	Flux (D) WY:97			110		280				170	2950		450		120	1330	12 200					2430	1880	1090		230 000	
	Flux (T) WY:97			690		3730				1300	10 700		1370			18 600	14 600					660 000	39 300	1 100 000		750 000	
	Solid (%) WY:97			83		93				87	72		67			93	17					100	95	100		69	
	Flux (S) WY:98	13·4	<7	540	580	3220	11	1230	430	1060	7240	1340	860	39	<67	16 100	2280	210	17	15	1	620 000	34 800	1 100 000	59 000	480 000	520 000
	Flux (D) WY:98			120		240				150	2830		430		110	1280	11 700					1480	1850	860		280 000	
	Flux (T) WY:98			660		3450				1210	10 100		1290			17 400	14 000					620 000	36 700	1 100 000		760 000	
	Solid (%) WY:98			82		93				87	72		67			93	16					100	95	100		63	
	Flux (S) WY:96-98	56·8	<28	2240	2410	13 400	45	5150	1790	4420	30 200	5600	3580	160	<280	67 200	9520	870	73	62	5	2 600 000	146 000	4 400 000	250 000	2 000 000	2 200 000
	Flux (D) WY:96-98			370		800				510	9470		1430		370	4290	38 700					8240	5670	3750		760 000	
	Flux (T) WY:96-98			2610	14 200					4940	39 700		5010			71 500	48 200					2 600 000	151 000	4 400 000		2 800 000	
	Solid (%) WY:96-98			86		94				90	76		71			94	20					100	96	100		73	
Ohio River at Cannelton	Flux (S) WY:96	40·8	<20	1470	1550	8150	24	3300	1020	2400	21 200	4480	2440	98	<204	37 000	5710	570	57	37	3	1 700 000	73 300	3 200 000	190 000	1 100 000	1 300 000
	Flux (D) WY:96			270		370		180		270	5120		830		260	4470	27 000					5310	700	1900		620 000	
	Flux (T) WY:96			1740		8520		3440		2680	26 300		3280			41 500	32 700					1 700 000	74 000	3 200 000		1 800 000	
	Solid (%) WY:96			84		96		95		90	81		75			89	17					100	99	100		65	
	Flux (S) WY:97	34·6	<17	1250	1320	6930	21	2770	870	2040	18 000	3810	2080	83	<173	48 500	4850	480	48	31	3	1 500 000	62 300	2 700 000	160 000	970 000	1 100 000
	Flux (D) WY:97			250		370		170		250	4740		770		240	4100	25 200					3190	610	1420		580 000	
	Flux (T) WY:97			1500		7300		2940		2290	22 800		2850			52 600	30 000					1 500 000	63 000	2 700 000		1 500 000	
	Solid (%) WY:97			83		95		94		89	79		73			92	16					100	99	100		63	

(*continued overleaf*)

Table III. (*continued*)

Location		S.S. (Mt)	Ag (t)	Cu (t)	Pb (t)	Zn (t)	Cd (t)	Cr (t)	Co (t)	Ni (t)	Ba (t)	V (t)	Li (t)	Be (t)	Mo (t)	P (t)	Sr (t)	As (t)	Sb (t)	Se (t)	Hg (t)	Fe (t)	Mn (t)	Al (t)	Ti (t)	TOC (t)	Total C (t)
	Flux (S) WY:98	17·3	<9	620	660	3450	10	1380	430	1020	8970	1900	1040	41	86	24 200	2420	240	24	16	1	720 000	31 000	1 300 000	81 100	480 000	550 000
	Flux (D) WY:98			220		320		160		220	4390		700		210	3630	22 900					1400	420	1050		510 000	
	Flux (T) WY:98			840		3770		1540		1230	13 400		1730			27 800	25 300					730 000	31 400	1 300 000		990 000	
	Solid (%) WY:98			74		91		89		83	67		60			87	10					100	99	100		49	
	Flux (S) WY:96-98	92·6	<46	3340	3520	18 500	56	7410	2320	5460	48 200	10 200	5560	220	<463	130 000	12 700	1300	130	83	7	3 900 000	167 000	7 200 000	440 000	2 600 000	3 000 000
	Flux (D) WY:96-98			740		1060		520		740	14 100		2300		710	12 200	75 000					9900	1740	4370		1 700 000	
	Flux (T) WY:96-98			4070		19 600		7930		6200	62 200		7860			142 000	88 000					3 900 000	169 000	7 200 000		4 300 000	
	Solid (%) WY:96-98			82		95		93		88	77		71			91	15					100	99	100		60	
Wabash River at New Harmony	Flux (S) WY:96	6·0	<3	150	160	840	4·2	390	78	230	2870	550	250	10	<30	7170	780	66	6	5	0·8	200 000	7170	380 000	22 700	140 000	190 000
	Flux (D) WY:96			55		22		46		42	1260		130		86	1020	4360	33				240	120	150		140 000	
	Flux (T) WY:96			210		860		440		280	4139		380			8190	5140	98				200 000	7300	380 000		290 000	
	Solid (%) WY:96			73		97		89		85	69		66			88	15	67				100	98	100		50	
	Flux (S) WY:97	6·7	<3	170	180	940	4·7	440	87	260	3230	620	280	11	<33	8070	870	74	7	5	0·9	220 000	8070	420 000	25 600	160 000	220 000
	Flux (D) WY:97			55		48		59		54	1370		140		93	2090	5190	32				380	110	180		150 000	
	Flux (T) WY:97			225		990		500		320	4600		420			10 200	6060	106				220 000	8180	420 000		310 000	
	Solid (%) WY:97			76		95		88		83	70		67			79	14	70				100	99	100		52	
	Flux (S) WY:98	6·7	<3	170	180	940	4·7	440	88	260	3230	620	280	11	<34	8080	880	74	7	5	0·9	220 000	8080	420 000	25 600	160 000	220 000
	Flux (D) WY:98			60		99		68		59	1380		140		93	2810	4860	36				270	61	170		150 000	
	Flux (T) WY:98			230		1040		510		320	4610		420			10 900	5730	110				220 000	8140	420 000		310 000	
	Solid (%) WY:98			74		90		87		82	70		67			74	15	68				100	99	100		52	
	Flux (S) WY:96-98	19·4	<10	486	510	2720	14	1260	253	760	9330	1790	820	31	<97	23 300	2530	210	19	16	3	640 000	23 300	1 200 000	73 800	470 000	620 000
	Flux (D) WY:96-98			165		170		170		160	4000		400		270	5920	14 400	100				890	300	500		440 000	
	Flux (T) WY:96-98			653		2890		1440		910	13 300		1220			29 200	16 900	310				640 000	23 600	1 200 000		910 000	
	Solid (%) WY:96-98			74		94		88		83	70		67			80	15	68				100	99	100		52	

Tennessee River at Paducuh	Flux (S) WY:96	1·1	<0·5	31	36	230	0·7	120	18	74	540	92	52	2	<5·3	2130	150	14	2	1	0·2	38 300	3190	68 100	4150		44 700
	Flux (D) WY:96			68		17		76			1130					1420	3440	50				1020	140	360		140 000	
	Flux (T) WY:96			99		250		200			1680					3550	3590	64				39 300	3330	68 400			
	Solid (%) WY:96			31		93		60			32					60	4	22				97	96	99			
	Flux (S) WY:97	1·4	0·7	40	47	310	1·0	150	24	97	710	120	68	2	<6·9	2780	190	18	2	1	0·2	50 000	4160	88 800	5410		50 300
	Flux (D) WY:97			84		100		93			1380					1710	4110	69				1370	170	450		160 000	
	Flux (T) WY:97			124		410		240			2090					4490	4300	87				51 400	4330	89 300			
	Solid (%) WY:97			32		75		62			34					62	5	21				97	96	99			
	Flux (S) WY:98	1·1	<0·5	31	36	230	0·7	120	18	74	540	91	52	2	<5	2120	150	14	2	1	0·2	38 100	3180	67 700	4130		44 500
	Flux (D) WY:98			67		330		75			1110					1240	3340	52				920	130	320		110 000	
	Flux (T) WY:98			98		560		190			1640					3360	3480	65				39 100	3310	68 000			
	Solid (%) WY:98			31		41		61			33					63	4	21				98	96	100			
	Flux (S) WY:96-98	3·5	<2	102	120	770	2·5	300	60	258	1790	300	170	6	<18	7020	490	46	6	3	0·6	126 000	10 500	225 000	13 700		150 000
	Flux (D) WY:96-98			220		450		240			3620					4300	10 900	170				3310	440	1130		410 000	
	Flux (T) WY:96-98			322		1220		630			5410					11 400	11 400	220				130 000	11 000	226 000			
	Solid (%) WY:96-98			32		63		61			33					62	4	21				97	96	99			
Ohio River near Grand Chain	Flux (S) WY:96	44·3	<22	1510	1600	5420	31	3500	930	2480	22 600	4520	2480	98	<222	62 100	5320	620	53	31	4	1 800 000	79 800	3 100 000	200 000	1 200 000	1 300 000
	Flux (D) WY:96			500		510		430		450	9660		1680		470	11 200	47 100	320				5250	1340	2490		960 000	
	Flux (T) WY:96			2000		8930		3900		2930	32 300		4160			73 300	52 400	940				1 800 000	81 200	3 100 000		2 200 000	
	Solid (%) WY:96			75		94		89		85	70		60			85	10	66				100	98	100		55	
	Flux (S) WY:97	53·0	<26	1800	1900	10 100	37	4190	1110	2970	27 000	5410	2970	120	<265	74 200	6360	740	64	37	4	2 100 000	95 400	3 800 000	240 000	1 400 000	1 500 000
	Flux (D) WY:97			520		730		480		470	9900		1750		480	12 900	46 000	340				5310	1130	2420		1 000 000	
	Flux (T) WY:97			2330		10 800		4670		3440	37 000		4720			87 100	52 400	1080				2 100 000	96 600	3 800 000		2 400 000	
	Solid (%) WY:97			78		93		90		86	73		63			85	12	69				100	99	100		58	

(continued overleaf)

Table III. (*continued*)

Location		S.S. (Mt)	Ag (t)	Cu (t)	Pb (t)	Zn (t)	Cd (t)	Cr (t)	Co (t)	Ni (t)	Ba (t)	V (t)	Li (t)	Be (t)	Mo (t)	P (t)	Sr (t)	As (t)	Sb (t)	Se (t)	Hg (t)	Fe (t)	Mn (t)	Al (t)	Ti (t)	TOC (t)	Total C (t)
	Flux (S) WY:98	36·1	<18	1230	1300	6860	25	2850	760	2020	18 400	3680	2020	79	<180	50 500	4300	510	43	25	3	1 400 000	65 000	2 600 000	160 000	940 000	10 000 000
	Flux (D) WY:98			440		850		470		400	8870		1530		430	9850	38 700	290				2650	630	1860		860 000	
	Flux (T) WY:98			1670		7710		3320		2430	27 300		3550			60 400	43 000	800				1 400 000	65 600	2 600 000		1 800 000	
	Solid (%) WY:98			73		89		86		83	67		57			84	10	63				100	99	100		52	
	Flux (S) WY:96-98	133	<66	4540	4800	25 400	93	10 500	2800	7470	68 100	13 600	7470	290	<667	190 000	16 000	1870	160	93	11	5 400 000	240 000	9 500 000	600 000	3 500 000	3 900 000
	Flux (D) WY:96-98			1460		2090		1370		1320	28 400		4950		1380	33 900	132 000	950				13 200	3100	6770		2 800 000	
	Flux (T) WY:96-98			6000		27 400		11 900		8790	96 500		12 400			220 000	148 000	2820				5 400 000	240 000	9 500 000		6 300 000	
	Solid (%) WY:96-98			76		92		88		85	71		60			85	11	66				100	99	100		55	
Mississippi River at Clinton	Flux (S) WY:96	4·7	<2	93	140	650	3	340	56	160	2610	360	140	6	<23	6520	650	37	5	5	0·4	140 000	9310	240 000	14 400	170 000	200 000
	Flux (D) WY:96			94		390		100		82	2160		360		56	5060	5030	48				5270	630	580		43 000	
	Flux (T) WY:96			190		1040		450		240	4770		500			11 600	5680	85				150 000	9940	240 000		600 000	
	Solid (%) WY:96			50		63		77		67	55		27			56	11	44				96	94	100		28	
	Flux (S) WY:97	5·1	<3	100	160	710	3	380	61	180	2850	390	150	6	<25	7120	710	41	6	5	0·4	150 000	10 200	260 000	15 800	180 000	220 000
	Flux (D) WY:97			96		390		130		82	2360		360		61	4910	5050	48				3680	630	300		370 000	
	Flux (T) WY:97			200		1110		510		260	5200		510			12 000	5760	89				160 000	10 800	260 000		550 000	
	Solid (%) WY:97			51		64		74		68	55		29			59	12	46				98	94	100		33	
	Flux (S) WY:98	3·6	<2	72	110	510	2	270	43	130	2020	280	110	4	<18	5060	510	29	4	4	0·3	110 000	7220	190 000	11 200	130 000	160 000
	Flux (D) WY:98			80		330		130		71	2090		310		57	3790	4390	44				1440	500	140		260 000	
	Flux (T) WY:98			150		840		400		200	4110		420			8840	4890	73				110 000	7720	190 000		390 000	
	Solid (%) WY:98			48		60		68		64	49		25			57	10	39				99	94	100		33	
	Flux (S) WY:96-98	13·4	<7	270	410	1870	8	990	160	470	7480	1030	390	16	<67	18 700	1870	110	15	13	1	400 000	26 700	690 000	41 400	480 000	590 000
	Flux (D) WY:96-98			270		1110		360		240	6610		1040		170	13 800	14 500	140				10 400	1750	1020		1 100 000	
	Flux (T) WY:96-98			540		2980		1350		700	14 100		1400			32 400	16 300	250				410 000	28 500	690 000		1 500 000	
	Solid (%) WY:96-98			50		63		73		67	53		27			58	11	43				97	94	100		31	

Missouri River near Culbert-son	Flux (S) WY:96	8·1	<4	140	120	630	2	540	81	310	6020	710	280	11	<41	4960	2200	110	17	3	0·2	200 000	2930	450 000	19 500	65 100	120 000
	Flux (D) WY:96			30				31		34	520		600		35	260	5510	27				230	50	26		140 000	
	Flux (T) WY:96			170				570		340	6540		880			520	7700	140				200 000	2980	450 000		210 000	
	Solid (%) WY:96			82				95		90	92		31			95	29	81				100	98	100		31	
	Flux (S) WY:97	9·9	<5	170	150	770	2	650	99	380	7330	860	340	13	<50	6040	2680	140	21	4	0·3	240 000	3570	540 000	23 800	79 300	150 000
	Flux (D) WY:97			28				29		25	450		570		33	180	5260	26				120	32	20		72 700	
	Flux (T) WY:97			200				680		400	7790		910			6220	7940	160				240 000	3600	540 000		150 000	
	Solid (%) WY:97			86				96		94	94		37			97	34	84				100	99	100		52	
	Flux (S) WY:98	4·6	<2	78	69	360	1	300	46	180	3410	400	160	6	<2·3	2810	1240	65	10	2	0·1	110 000	1660	250 000	11 100	36 900	69 100
	Flux (D) WY:98			25				28		17	380		520		30	59	4800	24				43	18	14		36 500	
	Flux (T) WY:98			100				330		190	3790		680			2870	6040	88				110 000	1680	250 000		73 300	
	Solid (%) WY:98			76				92		91	90		23			98	21	73				100	99	100		50	
	Flux (S) WY:96-98	22·7	<11	380	340	1770	5	1500	230	860	16 800	1970	770	29	<113	13 800	6120	320	48	9	0·7	540 000	8150	1 200 000	54 400	180 000	340 000
	Flux (D) WY:96-98			82				87		75	1360		1700		97	500	15 600	77				390	100	60		250 000	
	Flux (T) WY:96-98			470				1580		940	18 100		2470			14 300	21 700	400				540 000	8250	1 200 000		440 000	
	Solid (%) WY:96-98			82				94		92	93		31			97	28	80				100	99	100		42	
Yellow-stone River near Sydney	Flux (S) WY:96	21·7	<11	460	390	1760	7	1520	240	780	14 300	2020	740	33	<108	16 000	5640	260	26	13	0·9	610 000	11 700	1 300 000	62 900	240 000	370 000
	Flux (D) WY:96			30		24		18		19	564		360		21	260	5680	51		12		130	42	63		53 700	
	Flux (T) WY:96			480		1780		1540		800	14 900		1100			16 300	11 300	310		25		610 000	11 800	1 300 000		290 000	
	Solid (%) WY:96			94		99		99		98	96		67			98	50	84		52		100	100	100		82	
	Flux (S) WY:97	43·9	<22	920	790	3560	13	3080	480	1580	29 000	4090	1490	66	<220	32 500	11 400	530	53	26	2	1 200 000	23 700	2 700 000	130 000	480 000	750 000
	Flux (D) WY:97			36		29		29		24	670		400		25	340	6350	63		14		81	38	92		60 000	
	Flux (T) WY:97			960		3560		3100		1610	29 700		1890			32 800	17 800	590		40		1 200 000	23 800	2 700 000		540 000	
	Solid (%) WY:97			96		99		99		98	98		79			99	64	89		66		100	100	100		89	

(*continued overleaf*)

Table III. (*continued*)

Location		S.S. (Mt)	Ag (t)	Cu (t)	Pb (t)	Zn (t)	Cd (t)	Cr (t)	Co (t)	Ni (t)	Ba (t)	V (t)	Li (t)	Be (t)	Mo (t)	P (t)	Sr (t)	As (t)	Sb (t)	Se (t)	Hg (t)	Fe (t)	Mn (t)	Al (t)	Ti (t)	TOC (t)	Total C (t)
	Flux (S) WY:98	9·5	<5	200	170	760	3	660	100	340	6200	880	320	14	<47	6990	2460	110	11	6	0·4	260 000	5100	580 000	27 400	100 000	160 000
	Flux (D) WY:98			25		20		27		15	480		334		19	180	5130	42		11		15	24	38		33 100	
	Flux (T) WY:98			220		790		690		360	6710		650			7180	7590	160		16		260 000	5120	580 000		140 000	
	Solid (%) WY:98			89		97		96		96	93		49			97	32	73		35		100	100	100		76	
	Flux (S) WY:96-98	75·0	<38	1580	1350	6080	23	5250	830	2700	49 500	6900	2550	110	<375	55 500	19 500	900	90	45	3	2 100 000	40 500	4 700 000	220 000	820 000	1 300 000
	Flux (D) WY:96-98			91		73		74		59	1710		1090		65	780	17 200	160		36		226	100	193		150 000	
	Flux (T) WY:96-98			1670		6150		5330		2760	51 200		3640			56 300	36 700	1060		81		2 100 000	40 600	4 700 000		970 000	
	Solid (%) WY:96-98			95		99		99		98	97		70			99	53	85		56		100	100	100		85	
Missouri River at Omaha	Flux (S) WY:96	30·0	<15	390	480	1800	12	1590	270	1080	21 000	1800	690	30	150	19 500	7190	270	54	24	1	540 000	21 600	1 400 000	56 900	330 000	450 000
	Flux (D) WY:96			97		64		120		160	1350		2100		110	2100	23 150	96		220		370	3880	120		210 000	
	Flux (T) WY:96			490		1860		1710		1240	22 300		2790		260	21 600	30 300	360		250		540 000	25 400	1 400 000		540 000	
	Solid (%) WY:96			80		97		93		87	94		25		57	90	24	74		10		100	85	100		61	
	Flux (S) WY:97	35·6	<18	460	570	2140	14	1890	320	1280	25 000	2140	820	36	180	23 100	8540	320	64	28	1	640 000	25 600	1 600 000	67 600	390 000	530 000
	Flux (D) WY:97			110		78		150		160	2920		2370		150	2050	26 500	110		200		610	1540	180		280 000	
	Flux (T) WY:97			580		2210		2040		1440	27 800		3190		330	25 200	35 000	430		230		640 000	27 200	1 600 000		670 000	
	Solid (%) WY:97			80		96		93		89	90		26		55	92	24	74		12		100	94	100		59	
	Flux (S) WY:98	25·6	<13	660	410	1540	10	1360	230	920	17 900	1540	590	26	130	16 700	6140	230	46	20	1	460 000	18 400	1 200 000	48 600	280 000	380 000
	Flux (D) WY:98			78		53		100		99	2700		1850		120	1120	28 300	80		140		320	460	98		170 000	
	Flux (T) WY:98			740		1590		1460		1020	20 600		2430		240	17 800	26 500	310		160		460 000	18 900	1 200 000		450 000	
	Solid (%) WY:98			89		97		93		90	87		24		52	94	23	74		13		100	98	100		62	

	Flux (S) WY:96-98	91·1	<46	1510	1460	5470	36	4830	820	3280	63 800	5470	2100	91	<456	59 200	21 900	820	160	73	3	1 600 000	65 600	4 200 000	170 000	1 000 000	1 400 000
	Flux (D) WY:96-98			290		200		370		420	6980		6310		380	5270	70 000	290		570		1300	5880	410		650 000	
	Flux (T) WY:96-98			1800		5660		5200		3700	70 800		8410			64 500	91 800	1110		640		1 600 000	71 500	4 200 000		1 700 000	
	Solid (%) WY:96-98			84		97		93		89	90		25			92	24	74		11		100	92	100		61	
Missouri River at Hermann	Flux (S) WY:96	92·6	<46	1670	1850	7130	37	5290	930	3060	63 800	7410	2690	120	<463	70 400	19 500	830	130	74	3	2 100 000	67 600	5 000 000	230 000	1 400 000	1 700 000
	Flux (D) WY:96			190		140		150		230	8590		2900		280	6590	36 100	190		160		1010	330	710		360 000	
	Flux (T) WY:96			1860		7280		5340		3280	71 600		5580			77 000	55 500	1030		230		2 100 000	67 900	5 000 000		1 800 000	
	Solid (%) WY:96			90		98		97		93	88		48			91	35	81		32		100	100	100		79	
	Flux (S) WY:97	119	<59	2140	2380	9150	48	6660	1190	3920	80 800	9510	3450	160	<594	90 300	25 000	1070	170	95	4	2 700 000	86 800	6 400 000	300 000	1 800 000	2 100 000
	Flux (D) WY:97			210		210		210		260	10 000		2980		290	7890	42 000	220		170		2160	360	1110		450 000	
	Flux (T) WY:97			2350		9360		6860		4180	90 900		6420			98 200	67 000	1290		270		2 700 000	87 200	6 400 000		2 200 000	
	Solid (%) WY:97			91		98		97		94	89		54			92	37	83		36		100	100	100		80	
	Flux (S) WY:98	110	<55	1980	2190	8440	44	6140	1100	3620	74 500	8770	3180	140	<548	83 300	23 000	990	150	88	3	2 500 000	80 000	5 900 000	270 000	1 600 000	2 000 000
	Flux (D) WY:98			200		280		230		250	9710		2700		270	8450	41 400	210		170		3420	320	1380		420 000	
	Flux (T) WY:98			2180		8720		6360		3860	84 200		5880			91 700	64 400	1200		260		2 500 000	80 300	5 900 000		2 100 000	
	Solid (%) WY:98			91		97		96		94	88		54			91	36	82		34		100	100	100		79	
	Flux (S) WY:96-98	321	<161	5780	6420	24 700	130	18 000	3210	10 600	220 000	25 700	9310	420	<1606	240 000	67 400	2890	450	260	10	7 400 000	230 000	17 300 000	800 000	4 800 000	5 800 000
	Flux (D) WY:96-98			600		630		590		730	28 300		8580		840	22 900	120 000	620		500		6590	1020	3200		1 200 000	
	Flux (T) WY:96-98			6400		25 400		18 600		11 300	250 000		17 900			270 000	190 000	3520		750		7 400 000	240 000	17 300 000		6 100 000	
	Solid (%) WY:96-98			91		98		97		94	89		52			91	36	82		34		100	100	100		80	
Missi-ssippi River at Thebes	Flux (S) WY:96	74·6	<37	1570	1940	6940	37	4700	820	2840	45 500	6120	2310	97	<373	66 400	13 400	670	90	67	3	1 900 000	67 900	4 100 000	220 000	1 400 000	1 800 000
	Flux (D) WY:96			420		230		310		480	16 600		3240		430	20 900	50 000	340		230		4400	750	3280		1 000 000	
	Flux (T) WY:96			1990		7170		5010		3320	62 100		5560			87 300	63 500	1020		300		1 900 000	68 700	4 100 000		2 500 000	
	Solid (%) WY:96			79		97		94		85	73		42			76	21	66		22		100	99	100		57	

(continued overleaf)

Table III. (*continued*)

Location		S.S. (Mt)	Ag (t)	Cu (t)	Pb (t)	Zn (t)	Cd (t)	Cr (t)	Co (t)	Ni (t)	Ba (t)	V (t)	Li (t)	Be (t)	Mo (t)	P (t)	Sr (t)	As (t)	Sb (t)	Se (t)	Hg (t)	Fe (t)	Mn (t)	Al (t)	Ti (t)	TOC (t)	Total C (t)
	Flux (S) WY:97	80·0	<40	1670	2070	7410	40	5020	880	3030	48 600	6530	2470	100	<398	70 900	14 300	720	96	72	3	2 000 000	72 500	4 400 000	240 000	1 500 000	1 900 000
	Flux (D) WY:97			440		540		430		480	17 100		3320		480	21 600	52 700	360		250		7730	1060	4800		1 100 000	
	Flux (T) WY:97			2110		7950		5450		3510	65 700		5790			92 500	67 100	1080		320		2 000 000	73 600	4 400 000		2 600 000	
	Solid (%) WY:97			79		93		92		86	74		43			77	21	67		23		100	99	100		59	
	Flux (S) WY:98	94·0	<47	1970	2440	8740	47	5920	1030	3570	57 300	7710	2910	120	<470	83 700	16 900	850	110	85	4	2 300 000	85 500	5 200 000	280 000	1 800 000	2 300 000
	Flux (D) WY:98			470		1250		530		530	18 700		3400		530	23 400	55 200	420		270		7960	1070	5110		1 100 000	
	Flux (T) WY:98			2440		9990		6450		4100	76 000		6310			110 000	72 000	1270		350		2 300 000	86 600	5 200 000		2 900 000	
	Solid (%) WY:98			81		87		92		87	75		46			78	23	67		24		100	99	100		62	
	Flux (S) WY:96-98	248	<124	5210	6460	23 100	120	15 600	2730	9440	150 000	20 400	7700	320	<1241	220 000	44 700	2340	300	220	10	6 200 000	230 000	13 700 000	740 000	4 700 000	6 000 000
	Flux (D) WY:96-98			1330		2020		1280		1490	52 400		9970		1440	65 800	160 000	1120		740		20 100	2880	13 200		3 200 000	
	Flux (T) WY:96-98			6550		25 100		16 900		10 900	200 000		17 600			290 000	200 000	3360		970		6 200 000	230 000	13 700 000		7 900 000	
	Solid (%) WY:96-98			80		92		92		86	74		44			77	22	67		23		100	99	100		59	
Mississippi River at St. Francisville	Flux (S) WY:96	105	<52	2300	2510	11 500	52	6700	1360	4080	62 800	9010	3670	170	<524	98 500	15 700	1150	120	63	4	3 000 000	120 000	6 400 000	350 000	1 600 000	2 200 000
	Flux (D) WY:96			950		930		510		800	24 600		3430		740	27 100	77 100	530				5080	820	2420		1 700 000	
	Flux (T) WY:96			3250		12 430		7210		4880	87 400		7100			130 000	92 800	1680				3 000 000	120 000	6 400 000		3 300 000	
	Solid (%) WY:96			71		93		93		84	72		52			76	17	68				100	100	100		48	
	Flux (S) WY:97	143	<71	3140	3420	15 700	71	9130	1850	5560	85 600	12 300	4990	230	<713	130 000	21 400	1570	160	86	6	4 100 000	160 000	8 700 000	470 000	2 100 000	2 600 000
	Flux (D) WY:97			1230		1610		840		1090	31 400		3860		930	30 300	94 500	660				4840	1710	2680		2 400 000	
	Flux (T) WY:97			4370		17 300		9970		6650	120 000		8850			160 000	120 000	2230				4 100 000	160 000	8 700 000		4 500 000	
	Solid (%) WY:97			72		91		92		84	71		56			81	18	70				100	100	100		47	
	Flux (S) WY:98	124	<62	2730	2980	13 700	62	7940	1610	4840	74 500	10 700	4340	200	<621	120 000	18 600	1370	140	74	5	3 600 000	140 000	7 600 000	410 000	1 900 000	2 200 000
	Flux (D) WY:98			1090		1500		870		940	27 800		3960		810	28 600	85 300	590				1630	1210	2080		2 000 000	
	Flux (T) WY:98			3820		15 200		8810		5780	100 000		8320			150 000	100 000	1960				3 600 000	140 000	7 600 000		3 900 000	
	Solid % WY:98			71		90		90		84	75		52			80	19	70				100	100	100		49	

	Flux (S) WY:96-98	372	<186	8170	8920	40 900	190	23 800	4830	14 500	220 000	32 000	13 000	590	<1857	350 000	55 700	4090	410	220	14·9	10 800 000	410 000	22 700 000	1 200 000	5 600 000	6 700 000
	Flux (D) WY:96-98			3270		4040		2230		2840	83 800		11 300		2480	86 100	260 000	1780				11 600	3730	7180		6 200 000	
	Flux (T) WY:96-98			11440		44 900		26 000		17 300	300 000		24 300			440 000	320 000	5870				10 800 000	410 000	22 700 000		11 800 000	
	Solid % WY:96-98			71		91		92		84	73		53			80	17	70				100	100	100		47	
Atchafa-laya River at Melville	Flux (S) WY:96	52·0	<26	1030	1140	5170	21	3150	620	1910	28 900	4390	1860	83	<258	46 500	7230	520	46	26	3	1 400 000	51 700	3 000 000	170 000	720 000	830 000
	Flux (D) WY:96			380		630		220		340	11 000		1370		260	10 600	34 700	220				6030	1020	1630		810 000	
	Flux (T) WY:96			1420		5800		3370		2260	39 900		3230			57 100	42 000	740				1 400 000	52 700	3 000 000		1 500 000	
	Solid (%) WY:96			73		89		94		85	73		58			81	17	70				100	98	100		47	
	Flux (S) WY:97	79·0	<39	1570	1730	7850	31	4790	940	2900	43 900	6670	2820	130	<392	78 600	11 000	780	71	39	4	2 100 000	78 500	4 600 000	250 000	1 100 000	1 300 000
	Flux (D) WY:97			470		850		330		460	14 500		1540		300	11 900	42 700	290				7670	2520	1710		1 100 000	
	Flux (T) WY:97			2040		8700		5120		3360	58 500		4360			82 500	53 700	1070				2 100 000	81 000	4 600 000		2 200 000	
	Solid (%) WY:97			77		90		93		86	75		65			86	20	73				100	97	100		49	
	Flux (S) WY:98	66·0	<33	1320	1450	6580	26	4010	790	2440	36 800	5590	2370	110	<329	59 200	9200	660	59	33	3	1 800 000	65 800	3 900 000	210 000	920 000	1 100 000
	Flux (D) WY:98			430		740		360		400	12 500		1450		280	11 300	38 500	250				4830	1600	1100		970 000	
	Flux (T) WY:98			1740		7320		4380		2830	49 300		3820			70 500	47 700	910				1 800 000	67 400	3 900 000		2 000 000	
	Solid (%) WY:98			75		90		92		86	75		62			84	19	73				100	98	100		49	
	Flux (S) WY:96-98	197	<98	3920	4310	19 600	78	12 000	2350	7250	110 000	16 700	7050	310	<980	180 000	27 400	1960	180	98	10	5 300 000	200 000	11 600 000	630 000	2 700 000	3 100 000
	Flux (D) WY:96-98			1280		2220		910		1200	37 900		4360		830	33 800	120 000	760				18 500	5140	4430		2 900 000	
	Flux (T) WY:96-98			5200		21 800		12 900		8450	150 000		11 400			210 000	140 000	2710				5 300 000	204 000	11 600 000		5 700 000	
	Solid (%) WY:96-98			75		90		93		86	74		62			84	19	72				100	97	100		48	

Flux (S)[1] flux associated with suspended sediment
Flux (D)[2] flux associated with filtered water
Flux (T)[3] total flux (suspended + filtered water)
Solid (%)[4] percent of flux associated with suspended sediment

For purposes of the following discussion, the Mississippi Basin has been divided into three sections: upper, middle, and lower (Figure 1). The upper basin includes the Mississippi River at Clinton and at Thebes, as well as input from the Missouri River at Hermann. The middle basin begins at Thebes and includes inputs from the Ohio and the Arkansas Rivers and the diversion at the Old River Control Structure (Figure 1). The lower basin begins at the Mississippi River at St. Francisville, and includes the Atchafalaya River at Melville (Figure 1). The below average suspended sediment discharges for the study period seem to result from below average contributions from the upper rather than the middle or lower sections of the basin. The below average annual suspended sediment discharges for the Mississippi River as a whole (−17%) are similar to the below average fluxes for the upper Mississippi River based on the 3-year estimate for Thebes (−17%, Table IV; Meade, 1995). This holds because even though the 3-year average contributions for the Missouri River (Hermann) appear somewhat above average (+13%), those for the Mississippi River at Clinton are markedly below average (−50%, Table IV, Figure 1; Meade, 1995). Further, although flux contributions from the Ohio Basin were above average (+15%), estimates for the lower Mississippi River (St. Francisville) were below average by about the same amount noted for Thebes (−17%; Table IV). Finally, estimates for the Atchafalaya (which receives, on average, 25% of Mississippi River water diverted at the Old River Control Structure; Meade, 1995) also seem to be about 17% below average (Table IV). Hence, there seems to have been no net change in suspended sediment fluxes in the middle and lower sections of the Mississippi Basin (between Thebes and St. Francisville including the Atchafalaya). Thus, the lower than average suspended sediment discharges from the Mississippi River Basin to the Gulf of Mexico during the study period probably resulted from below average suspended sediment contributions from the upper section of the basin.

Average dissolved trace element concentrations for the Mississippi River determined under the current NASQAN programme are markedly lower than those reported during the historical NASQAN programme (Table V), and are similar to but slightly higher than those reported by Shiller and Boyle (1987), Garbarino *et al*. (1995), and Taylor and Shiller (1997). This is likely to reflect the required use of 'clean' sampling, processing, and analytical techniques in the NASQAN programme rather than an overall improvement in water quality in the basin (e.g. Horowitz *et al*., 1994, 1996a,b). As noted in previous publications, the slightly higher values reported herein for several elements probably result from sample processing artifacts rather than sample contamination (Horowitz *et al*., 1996a,b).

Annual suspended sediment-associated trace element fluxes, as well as those for phosphorus (P) and total organic carbon (TOC), have been calculated for the Mississippi River and its major tributaries (Table III). Further, where data were available, total trace element fluxes (dissolved plus suspended sediment-associated) also have been calculated (Table III). Finally, where appropriate, the percentages of solid-phase contributions to total trace element fluxes have been summarized (Table VI). Note that the majority (typically $\geq$75%) of the Cu, Zn, Cr, Ni, Ba, P, As, Fe, Mn, and Al are transported in association with suspended sediment

Table IV. Summary of estimated annual suspended sediment fluxes for the 1996, 1997, and 1998 water years compared to long term averages

Sample location	Long-term average[a] ($\times 10^6$ t year^{-1})[b]	This study ($\times 10^6$ t year^{-1})[b]	Difference (%)
Mississippi River at Clinton	9	4·5	−50
Missouri at Hermann	95	107	13
Mississippi River at Thebes	100	83	−17
Ohio River near Grand Chain	40	46	15
Mississippi River at St. Francisville	150	124	−17
Atchafalaya River at Melville	80	66	−17
Mississippi River Basin	230	190	−17

[a] Meade (1995).
[b] Megatonnes.

Table V. Comparison of Mississippi River dissolved trace element concentrations from a variety of sources

Trace element	Unit	Historical NASQAN[a]	Shiller and Boyle[b]	Shiller[c]	Current NASQAN[d]
Cadmium	ng l^{-1}	2800	13	9–23	<500
Chromium	ng l^{-1}	1150	73		1100
Copper	ng l^{-1}	5100	1500	950–1900	2100
Nickel	ng l^{-1}	1800	1400	700–2100	1800
Lead	ng l^{-1}	2100		4–19	<500
Zinc	ng l^{-1}	6400	200	130–460	2200
Cobalt	ng l^{-1}	<3000			<500
Molybdenum	ng l^{-1}	<10 000	1100	770–3800	1800
Vanadium	ng l^{-1}	<6000	1200	260–1800	<3000
Arsenic	ng l^{-1}	1800			1100
Barium	ng l^{-1}	69 000		48 000–82 000	56 000
Iron	μg l^{-1}	45	1·7	0·5–4·5	6·7
Manganese	μg l^{-1}	11		0·05–3·8	1·8

[a] Pre-1991 NASQAN programme.
[b] Shiller and Boyle (1987).
[c] Shiller (1997) ranges for the lower Mississippi River.
[d] Averages for the 1996, 1997, and 1998 water years for the lower Mississippi River.

(Table VI). This is in line with results for other rivers reported previously by Horowitz (1995), and supports the contention of Windom *et al*. (1991) and Horowitz (1995), among many others, that the majority of fluvial trace element transport is dominated by the solid phase. On the other hand, Sr fluxes seem to be dominated by the dissolved fraction, whereas the movement of Li and TOC seems to be about equally divided between both phases (Table VI).

Although trace element fluxes increase downstream in the Ohio River Basin, the solid-phase contributions to those fluxes decrease (Tables III and VI). This could be the result of sediment trapping behind dams such as at Greenup and Cannelton. On the other hand, this is belied by the overall increase in suspended sediment fluxes in the same direction (Table III). In fact, the reduction in solid-phase-associated trace element fluxes in the Ohio River seems to be the result of two forms of dilution. Note the lower average suspended sediment-associated concentrations between Greenup and Cannelton for parameters such as Cu, Zn, Cd, Cr, etc. (Table II). Further, note the markedly lower sediment-associated fluxes from the Wabash and particularly the Tennessee Rivers (Table VI). Hence, it would seem that the downstream reduction of sediment-associated fluxes in the Ohio River results from (1) the downstream addition of suspended sediment containing lower trace element concentrations, and/or (2) the addition of greater dissolved trace element contributions.

As with trace elements, P and TOC fluxes increase, whereas the solid-phase contributions for both decrease, downstream in the Ohio River Basin (Table VI). The pattern for TOC appears similar to that for the trace elements cited above; however, the picture is incomplete due to a lack of sediment-associated TOC data for the Tennessee River (Table II). The pattern for P seems to be markedly different from the others. Sediment-associated P concentrations actually increase downstream in the Ohio River. Further, note the markedly higher sediment-associated contributions from the Tennessee River relative to the mainstem Ohio (Table VI). Similarly, dissolved P contributions also increase downstream in the Ohio River; however, the rate of increase for dissolved P is markedly higher than that for sediment-associated P (Table III). Hence, the reduction in solid-phase-associated P contributions to the overall P flux in the Ohio River would seem to be the result of markedly greater increases in the amounts of dissolved P relative to sediment-associated P. This may be a reflection of land-use changes (increased agricultural activity), as well as increases in population density.

Despite the presence of numerous dams and their associated impoundments, which have substantially reduced suspended sediment fluxes within the Missouri River Basin, as well as contributions to the upper

Table VI. Summary of the percentage of solid-phase contributions to total trace element fluxes in the four NASQAN basins

Basin	Subbasin	Site	Cu	Zn	Cr	Ni	Ba	Li	P	Sr	As	Se	Fe	Mn	Al	TOC
Mississippi		Mississippi at Clinton	50	63	73	67	53	27	58	11	43		97	94	>99	55
		Mississippi at Thebes	80	92	92	86	74	44	77	22	67		>99	99	>99	59
		Mississippi at St. Francisville	71	91	91	84	73	54	80	18	70		>99	99	>99	47
	Missouri	Missouri near Culbertson	82		94	92	93	31	97	28	80		>99	99	>99	42
	Missouri	Missouri at Omaha	84	97	93	89	90	25	92	24	74		>99	99	>99	61
	Missouri	Missouri at Hermann	91	98	97	94	89	52	91	36	82		>99	>99	>99	80
	Missouri	Yellowstone near Sydney	95	99	99	98	97	70	99	53	85		>99	>99	>99	85
	Ohio	Ohio at Greenup	86	94		90	76	71	94	20			>99	96	>99	73
	Ohio	Ohio at Cannelton	82	95	93	88	77	71	91	15			>99	96	>99	60
	Ohio	Ohio near Grand Chain	76	92	88	85	71	60	85	11	66		>99	99	>99	55
	Ohio	Wabash at New Harmony	74	94	88	83	70	67	80	15	68		>99	99	>99	52
	Ohio	Tennessee at Paducah	32	63	61		33		62	4	21		>99	96	>99	
	Atchafalaya	Atchafalaya at Melville	75	90	93	86	74	62	84	19	72		>99	97	>99	48
Columbia		Columbia at Northport	79	90	41		19		55	3			99	91	98	8
		Columbia at Vernita	19	52	39		10		58	2			94	84	97	10
		Columbia at Warrendale	44	74	54		38	13	61	7	17		99	95	>99	26
		Columbia near Beaver Army Terminal	62	81	67		59		73	16	24		99	95	>99	33
	Snake	Snake River at Burbank	40	73	50	33	46	10	53	5	14		97	93	98	25
	Willamette	Willamette at Portland	82	89			91		90	50			99	94	>99	33
Rio Grande		Rio Grande at El Paso	84	92	92	87	80	18	90	21	44		>99	99	>99	56
		Rio Grande at Foster Ranch	97	99	99	98	92	72	>99	66	95		>99	>99	>99	94
		Rio Grande at Laredo			78	72	42		59	9			>99	96		51
		Rio Grande near Brownsville	57	84	73	61	33	11	61	6	26		>99	99	>99	42
	Arroyo Colorado	Arroyo Colorado at Harlingen	47	81	76	54	46	12	43	8	17	10	>99	97	>99	42
Colorado		Colorado near Cisco	87	97	96	94	85	57	96	35	84	36	>99	99	>99	59
		Colorado above Diamond Creek	65	95	94	90	75	33	64	12	67	7	>99	99	>99	82
		Colorado at NIB		81	87	85	52	11	94	9	40	7	>99	97	>99	46
	Green River	Green River at Green River	91	98	98	96	91	69	98	41	85	33	>99	>99	>99	59
	San Juan	San Juan near Bluff	96	99	99	99	97	84	97	62		56	>99	>99	>99	52

Mississippi River relative to historic levels (Meade, 1995), there is slightly over a three-fold downstream increase between the upper section of the basin (Culbertson) and its mouth at Hermann (Table III, Figure 1). It should be noted that if the estimated contributions from the Yellowstone River are added to those of the upper Missouri (Culbertson), then there are essentially no net changes in suspended sediment fluxes between the upper and middle sections of the basin. Hence, the observed three-fold net increase in suspended sediment fluxes in the Missouri River probably can be attributed to contributions from the lower basin between Omaha and Hermann (Table III, Figure 1). Although this is arithmetically correct, the estimated errors for the Yellowstone at Sydney site are the largest and most negative (−23%) for all the NASQAN sediment chemical sites; further, this error is unlikely to be balanced by the slightly positive estimated error (8%) for the Culbertson site (Horowitz *et al*., 2001). Although there seems to be no net change in suspended sediment fluxes between the upper and middle section of the Missouri River, this should not be viewed as an indication that sediment deposition and resuspension are not occurring in this section of the basin. This view is supported by the significant chemical differences in suspended sediment-associated trace element concentrations (Table II) for the various sites (Horowitz *et al*., 2001).

Not surprisingly, because the sediment-associated contributions to trace element and P fluxes average more than 85%, trace element and P fluxes in the Missouri River basin tend to mirror those for suspended sediment (Tables III and VI). The downstream pattern for TOC is similar to that for trace elements and P; however, unlike the other parameters, there is a significant downstream increase (from 42 to 85%) in the contribution of sediment-associated TOC to total TOC fluxes (Tables III and VI). This may be a reflection of downstream increases in the drainage area of the basin, but is more likely the result of changes in land-use/socioeconomic factors such as increased agricultural activity and population density.

The downstream fluxes of suspended sediment, trace elements, P, and TOC significantly increase in the Mississippi River Basin (Table III). These increases are markedly greater between the upper section (Clinton to Thebes) of the basin relative to the middle section (Thebes to St. Francisville) of the basin (Table III). Although there are differences from one parameter to another, the increases between the upper and middle basin average about 20-fold whereas those between the middle and lower basin average about 1·5-fold. There also is a marked change in the relative significance of solid-phase flux contributions between the upper and middle of the basin (Table VI). In the former, the increases average around 40%, whereas there is relatively little change between the latter. Not surprisingly, this seems to be a reflection of a combination of the flow regime (e.g. contributions from the Missouri River upstream of Thebes) and a lack of substantial sediment storage in the upper part of the Mississippi River Basin, in conjunction with the flow regime (e.g. contributions from the Ohio and Arkansas Rivers, dilution due to more water) and the effects of substantial sediment storage and resuspension within the middle section of the basin (Meade, 1995). In effect, by the time upper Mississippi River-derived suspended sediment reaches St. Francisville, there has been sufficient mixing of new and stored sediment to mask any contributions from the upper Mississippi, the Missouri, the Ohio, and the Arkansas Rivers. This interpretation may be skewed by the lack of mainstem sampling sites between Thebes and St. Francisville, a distance of some 1100 km, which precludes an assessment of the downstream impact (distance) of contributions from the Ohio and Arkansas River Basins on the Mississippi River, as well as any opportunity to identify potential sources and/or sinks for suspended sediment in the middle section of the river. However, the effect of sediment storage and release on sediment-associated trace element concentrations and fluxes in the Mississippi River Basin might be inferred from the very similar below average annual estimates for suspended sediment fluxes (−17%), as well as the relative similarity in suspended sediment-associated contributions to total trace element fluxes between Thebes and St. Francisville (the middle section of the basin) relative to the observed differences between Clinton and Thebes (the upper section of the basin) (Table VI).

There is significant interannual variability in the suspended sediment-associated chemical fluxes along the mainstem Mississippi River and from its major tributaries (Table III). This occurs despite the fact that site-specific interannual sediment-associated chemical concentrations typically vary less than the analytical errors associated with the methods used to determine them (Horowitz *et al*., 2001). Hence, the interannual variations in chemical fluxes in the basin (Table III) predominantly result from substantial year-to-year differences in discharge and the concentration of suspended sediment itself, rather than from year-to-year sediment-associated chemical differences. This has been noted before in other, though markedly smaller rivers than the Mississippi (Horowitz, 1995). These results, along with those discussed in the previous paragraph, would imply that the effects of any significant annual changes in solid-phase-dominated chemical concentrations on chemical fluxes from the upper section of the Mississippi River Basin probably could be masked by annual variations in discharge and suspended sediment concentrations throughout the basin, as well as by sediment storage and resuspension processes in the middle section of the basin. Thus, it probably will take a substantial amount of travel time before chemical concentration changes in the upper section of the Mississippi River can be detected in the chemical concentrations and/or fluxes in the lower section of the basin or in discharges to the Gulf of Mexico.

THE COLUMBIA RIVER BASIN

Basic geographic data covering the Columbia River and its major tributaries are provided in Table I, and a schematic of the sampling sites is provided in Figure 2. Mean and median chemical concentrations for suspended sediment-associated trace elements from the basin are provided in Table II. For purposes of the following discussion, the Columbia River, like the Mississippi River, has been divided into three sections. The upper basin includes the Northport and Vernita sites and extends to, but does not include the Snake River (Figure 2). The middle portion of the basin includes input from the Snake River and extends through Warrendale (Figure 2). Finally, the lower portion of the basin begins where the Willamette River enters the Columbia River, and includes the Beaver Army Terminal site (Figure 2).

The sediment-associated chemical concentrations for Ag, Cu, Pb, Zn, Cd, Cr, Co, Ba, Mo, Sb, Hg, and Fe for the Northport site are markedly higher than typical background levels for fine-grained sediment; Zn is nearly 200 times greater, whereas Cu and Sb are nearly 20 times greater, Pb, Ag, Cd, and Mo are nearly 10 times greater, and Hg is about two times greater than background (Table II; Horowitz, 1991; Persaud *et al.*, 1993). These elevated concentrations probably are related to the release of mining-related materials upstream of the US/Canadian border (Bortleson *et al.*, 1994). Note that despite the presence of seven dams, including the Grand Coulee, at least some of these elevated levels (Cu, Pb, Zn, and Cd) appear

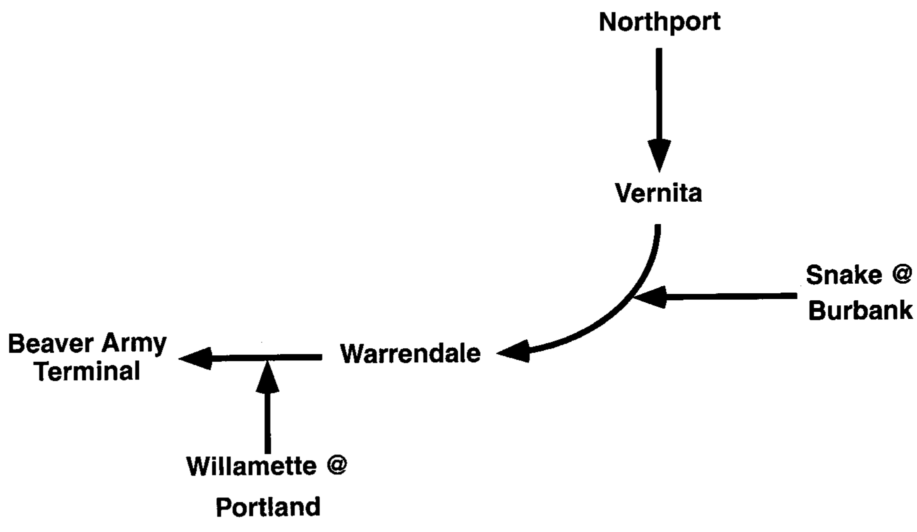

Figure 2. A schematic diagram of the NASQAN Columbia River Basin sampling sites (not to scale). The arrows indicate downstream flow direction

to extend to the Vernita site, some 550 km downstream from Northport (Table II). Some of the elevated sediment-associated trace elements detected at Vernita may not have originated at Northport. Mining-related, trace element-rich suspended sediment also may have come from both the Spokane River (about 160 km downstream from Northport) and the Okanogan River (about 320 km downstream from Northport), which receive discharges from the Bunker Hill and the Silver Mountain Mine Superfund sites, respectively. Hence, the sediment geochemistry of the upper Columbia River Basin is substantially affected by mining or mining-related activities. In fact, despite substantive discharges from the Snake River, it may be possible, based on elevated Zn levels, that the chemical impacts noted at Northport and Vernita may even extend as far downstream as Warrendale (Table II).

Suspended sediment fluxes to the Pacific Ocean (Columbia near Beaver Army Terminal) for the study period averaged about 12·5 Mt. This is about 25% greater than the annual average (Table I). However, the use of average values for the study period is misleading because, based on long-term water and suspended sediment discharge records, none of the three water years seem to have been typical (Tables I and VII). In the upper basin, sediment fluxes for the 1996 and 1997 water years were similar (about 0·7 Mt); however, in 1998, fluxes were about 20% lower (Table VII). Further, during the 1998 water year, suspended sediment fluxes from the Snake River (about 1 Mt), as well as those estimated for Warrendale, were about 50% below average (Tables I and VII). Finally, during the 1996 and 1997 water years, suspended sediment fluxes from the Willamette were 1000% and 600% greater than normal, whereas during the 1998 water year fluxes were 25% below normal (Tables I and VII). In fact, the highest discharges and suspended sediment concentrations on record were measured at the Portland site in February 1996 (Kelly and Hooper, 2001). These conditions combined to produce higher than average suspended sediment fluxes (about 16·5 Mt) at the mouth of the Columbia River during 1996 and 1997, and a markedly lower than average flux (4·5 Mt) in 1998.

With the exception of the upper Columbia River Basin, and despite the presence of numerous dams and reservoirs downstream throughout the basin, total net suspended sediment fluxes for 1996, 1997, and 1998 seem to display a fairly simple additive pattern (Table VII). Between Northport and Vernita, suspended sediment fluxes remained relatively constant (about 2 Mt). However, based on the sharp reduction in both trace element concentrations and fluxes between Northport and Vernita (Table VII), substantial deposition and remobilization (sediment mixing) took place. In other words, trace element-rich suspended sediment from Northport, and despite potential contributions from the Spokane and Okanogan Rivers, seems to have been diluted with less trace element-rich material in a downstream direction. During this period, there was a 7·5-fold increase in suspended sediment fluxes between Vernita (about 2 Mt) and Warrendale (about 15 Mt). About 40% of this increase can be attributed to inputs from the Snake River (about 5 Mt). The remaining increases probably are attributable to inflows from the Yakima, John Day, and Deschutes River Basins, especially the latter two which are heavily agricultural (cropping and particularly grazing). Between Warrendale and Beaver Army Terminal (about 38 Mt) there was a further 2·5-fold increase in suspended sediment fluxes. About 78% of this increase can be attributed to inputs from the Willamette River (Table VII). The remaining increases probably are the result of inputs from the Cowlitz and Toutle Rivers, which join the Columbia River downstream from Portland.

In the Columbia River Basin, as in other river systems, substantial quantities of trace elements are transported in association with suspended sediment (Table VI) (Windom *et al*., 1991; Horowitz, 1995; this study). However, there are marked variations throughout the system in response to significant changes in chemical levels and suspended sediment concentration (Tables II, VI and VII). If the Northport site is excluded, where the results are heavily skewed by the presence of mining-related discharges, there is a steady increase in sediment-associated trace element transport, reflecting the increase in sediment transport in general, as water moves downstream from the upper Columbia to its mouth (Tables VI and VII). This seems to be more in response to tributary inflows rather than to changes in the mainstem itself. Note the marked increases in transport of sediment and sediment-associated trace elements between Vernita and Warrendale after the Snake River enters the mainstem Columbia River, as well as the further increases

Table VII. Summary of estimated Columbia Basin fluxes for the 1996, 1997, and 1998 (see footnotes for Table III)

Location		S.S. (Mt)	Ag (t)	Cu (t)	Pb (t)	Zn (t)	Cd (t)	Cr (t)	Co (t)	Ni (t)	Ba (t)	V (t)	Li (t)	Be (t)	Mo (t)	P (t)	Sr (t)	As (t)	Sb (t)	Se (t)	Hg (t)	Fe (t)	Mn (t)	Al (t)	Ti (t)	TOC (t)	Tot. C (t)
Columbia River at Northport	Flux (S) WY:96	0·7	3	610	160	3690	2	97	29	33	840	53	22	1	15	900	270	8	48	1	0·2	53 600	1390	37 600	1600	13 900	20 900
	Flux (D) WY:96			170		420		130			3630					770	10 100					830	120	960		160 000	
	Flux (T) WY:96			780		4100		230			4460					1670	10 400					54 400	1510	38 500		180 000	
	Solid (%) WY:96			78		90		43			19					54	3					98	92	98		8	
	Flux (S) WY:97	0·7	3	620	160	3710	2	98	29	34	840	53	22	1	15	910	270	8	48	1	0·2	53 900	1400	37 800	1610	14 000	21 000
	Flux (D) WY:97			160		420		140			3650					580	9940					780	140	1020		170 000	
	Flux (T) WY:97			780		4140		240			4490					1490	10 200					54 700	1540	38 800		180 000	
	Solid (%) WY:97			79		90		41			19					61	3					99	91	97		8	
	Flux (S) WY:98	0·5	2	470	120	2850	2	75	23	26	650	41	17	1	12	700	210	6	37	1	0·1	41 400	1080	29 100	1240	10 800	16 100
	Flux (D) WY:98			110		300		120			2780					670	8100					620	110	680		120 000	
	Flux (T) WY:98			580		3160		190			3420					1370	8310					42 100	1180	29 700		130 000	
	Solid (%) WY:98			81		90		40			19					51	3					99	91	98		8	
	Flux (S) WY:96-98	1·9	9	1700	440	10 300	7	270	81	93	2320	150	60	3	43	2520	750	22	130	2	0·4	150 000	3870	100 000	4450	38 700	58 000
	Flux (D) WY:96-98			450		1140		390			10 100					2020	28 200					2230	370	2260		450 000	
	Flux (T) WY:96-98			2150		11 400		660			12 400					4540	28 900					150 000	4240	110 000		490 000	
	Solid (%) WY:96-98			79		90		41			19					55	3					99	91	98		8	
Columbia River at Vernita Bridge	Flux (S) WY:96	0·7	<1	54	81	420	2	120	15	88	480	71	22	1	<3·7	1180	210	12	2	1		30 200	1400	43 400	2570	30 200	40 400
	Flux (D) WY:96			220		490		180			4420					730	12 400					1970	270	1550		280 000	
	Flux (T) WY:96			280		910		300			4900					1910	12 600					32 100	1660	45 000		310 000	
	Solid (%) WY:96			20		46		39			10					62	2					94	84	97		10	
	Flux (S) WY:97	0·7	<1	55	82	420	2	120	15	89	490	72	22	1	<3·7	1190	210	12	2	1		30 400	1410	43 700	2590	30 400	40 800
	Flux (D) WY:97			230		370		180			4200					910	12 400					1830	280	1730		290 000	
	Flux (T) WY:97			280		790		300			4690					2090	12 600					32 200	1680	45 500		320 000	
	Solid (%) WY:97			20		53		39			10					57	2					94	84	96		10	

(*continued overleaf*)

Table VII. (*continued*)

Location		S.S. (Mt)	Ag (t)	Cu (t)	Pb (t)	Zn (t)	Cd (t)	Cr (t)	Co (t)	Ni (t)	Ba (t)	V (t)	Li (t)	Be (t)	Mo (t)	P (t)	Sr (t)	As (t)	Sb (t)	Se (t)	Hg (t)	Fe (t)	Mn (t)	Al (t)	Ti (t)	TOC (t)	Tot. C (t)
	Flux (S) WY:98	0·5	<1	41	61	320	1	89	11	67	370	54	17	1	<2·8	890	160	9	1	1		22 800	1050	32 700	1940	22 700	30 500
	Flux (D) WY:98			190		190		140			2970					720	10 100					1120	180	830		170 000	
	Flux (T) WY:98			230		500		230			3330					1610	10 300					23 800	1230	33 500		190 000	
	Solid (%) WY:98			18		63		39			11					55	2					95	86	98		12	
	Flux (S) WY:96-98	2·0	<1	150	220	1160	5	320	41	240	1340	200	61	3	<10	3250	570	34	5	2		83 300	3860	120 000	7110	83 300	110 000
	Flux (D) WY:96-98			640		1050		510			11 600					2360	34 900					4920	720	4110		730 000	
	Flux (T) WY:96-98			790		2210		830			12 900					5610	35 500					88 200	4580	120 000		820 000	
	Solid (%) WY:96-98			19		52		39			10					58	2					94	84	97		10	
Snake River at Burbank	Flux (S) WY:96	1·7	<1	82	40	220	1	90	37	58	1080	200	62	3	<8·3	2330	370	18	2	1	0·1	83 400	2330	130 000	9000	53 300	53 300
	Flux (D) WY:96			92		82		95		160	1280		490		92	2150	6120	120				3550	230	3600		180 000	
	Flux (T) WY:96			170		300		180		220	2360		552			4480	6490	140				87 000	2560	130 000		230 000	
	Solid (%) WY:96			48		73		50		27	46		11			52	6	13				96	91	100		23	
	Flux (S) WY:97	2·2	<1	110	53	290	1	120	49	78	1450	270	82	4	<11·1	3110	490	24	3	1	0·2	110 000	3110	170 000	12 000	71 200	71 200
	Flux (D) WY:97			140		93		100		120	1490		620		100	2440	7370	130				3130	220	2280		190 000	
	Flux (T) WY:97			250		380		220		200	2940		700			5550	7860	150				110 000	3330	170 000		260 000	
	Solid (%) WY:97			44		76		55		39	49		12			56	6	16				100	93	100		27	
	Flux (S) WY:98	1·1	<1	53	26	140	0	59	24	38	710	130	40	2	<5·4	1520	240	12	1	1	0·1	54 400	1520	81 700	5880	34 800	34 800
	Flux (D) WY:98			130		63		75		64	1100		520		69	1540	5650	88				1250	86	610		110 000	
	Flux (T) WY:98			180		200		130		100	1810		560			3060	5890	100				55 600	1610	82 300		140 000	
	Solid (%) WY:98			30		70		45		38	39		7			50	4	12				98	94	99		25	
	Flux (S) WY:96-98	5·0	<3	240	120	650	2	270	110	170	3240	600	180	9	<25	6970	1100	55	6	3	0·4	250 000	6970	370 000	26 900	160 000	160 000
	Flux (D) WY:96-98			360		240		270		350	3880		1630		260	6130	19 300	330				7930	540	6480		490 000	
	Flux (T) WY:96-98			600		890		540		520	7120		1810			13 100	20 400	390				260 000	7510	380 000		650 000	
	Solid (%) WY:96-98			40		73		50		33	46		10			53	5	14				96	93	97		25	

Columbia River at Warrendale	Flux (S) WY:96	5·5	<3	270	150	1210	7	340	100	220	1370	660	170	9	28	7170	1490	50	8	3	0·6	260 000	6620	400 000	28 100	150 000	180 000
	Flux (D) WY:96			340		420		280			5560		1040			4710	20 200	240				2190	560	760		440 000	
	Flux (T) WY:96			610		1630		620			8920		1210			11 900	21 600	280				270 000	7180	400 000		590 000	
	Solid (%) WY:96			45		74		55			38		14			60	7	17				99	92	100		26	
	Flux (S) WY:97	6·7	<3	330	180	1470	9	410	130	270	4060	800	200	11	33	8660	1800	60	10	3	0·7	320 000	8000	480 000	34 000	190 000	220 000
	Flux (D) WY:97			360		450		290			5400		1080			4760	20 600	250				1900	310	990		480 000	
	Flux T WY:97			690		1920		700			9470		1280			13 400	22 400	310				320 000	8310	480 000		660 000	
	Solid (%) WY:97			47		76		59			43		16			65	8	19				99	96	100		28	
	Flux (S) WY:98	3·0	<2	140	80	650	4	180	56	120	1800	360	89	5	15	3840	800	27	4	1	0·3	140 000	3550	210 000	15 100	82 800	97 500
	Flux (D) WY:98			230		300		210			3930		830			3210	16 400	170				650	88	420		300 000	
	Flux (T) WY:98			370		950		400			5740		920			7050	17 200	200				140 000	3640	210 000		380 000	
	Solid (%) WY:98			39		68		46			31		10			55	5	13				100	98	100		22	
	Flux (S) WY:96-98	15·1	<8	740	410	3330	20	940	290	610	9230	1820	450	24	76	19 700	4090	140	23	8	2	730 000	10 200	1 100 000	77 200	420 000	500 000
	Flux (D) WY:96-98			930		1180		780			14 900		2950			12 700	57 100	660				4730	960	2160		1 200 000	
	Flux (T) WY:96-98			1670		4510		1720			24 100		3410			32 400	61 200	790				730 000	19 100	1 100 000		1 600 000	
	Solid (%) WY:96-98			44		74		54			38		13			61	7	17				99	95	100		26	
Willamette River at Portland	Flux (S) WY:96	10·1	<5	570	230	1440	3	800	250	460	4650	1620	260	15	<51	18 200	2530	81	10	4	0·8	600 000	18 200	840 000	68 800	350 000	400 000
	Flux (D) WY:96			81		140					280					1290	1540					4440	770	4110		96 300	
	Flux (T) WY:96			650		1570					4940					19 500	4070					600 000	19 000	840 000		450 000	
	Solid (%) WY:96			87		91					94					93	62					99	96	100		79	
	Flux (S) WY:97	6·7	<3	380	150	950	2	530	170	310	3090	1080	180	10	<34	12 100	1680	54	7	3	0·5	400 000	12 100	560 000	45 700	240 000	270 000
	Flux (D) WY:97			84		110					290					1380	1620					3770	780	2330		100 000	
	Flux (T) WY:97			460		1060					3380					13 500	3300					400 000	12 900	560 000		340 000	
	Solid (%) WY:97			82		90					91					90	51					99	94	100		69	

(*continued overleaf*)

Table VII. (*continued*)

Location		S.S. (Mt)	Ag (t)	Cu (t)	Pb (t)	Zn (t)	Cd (t)	Cr (t)	Co (t)	Ni (t)	Ba (t)	V (t)	Li (t)	Be (t)	Mo (t)	P (t)	Sr (t)	As (t)	Sb (t)	Se (t)	Hg (t)	Fe (t)	Mn (t)	Al (t)	Ti (t)	TOC (t)	Tot. C (t)
	Flux (S) WY:98	0·8	<1	42	17	110	0·2	60	19	35	350	120	20	1	<4	1360	190	6	1	0	0·1	44 800	1360	62 900	5160	26 500	30 300
	Flux (D) WY:98			49		63					180					940	1200					1330	400	370		54 800	
	Flux (T) WY:98			91		170					530					2310	1390					46 100	1770	63 300		81 300	
	Solid (%) WY:98			47		63					65					59	14					97	77	99		33	
	Flux (S) WY:96-98	17·6	<9	980	410	2500	5	1390	440	810	8090	2820	460	26	<88	31 700	4400	140	18	7	1	1 000 000	31 700	1 500 000	120 000	620 000	700 000
	Flux (D) WY:96-98			210		300					760					3610	4360					9550	1950	6820		250 000	
	Flux (T) WY:96-98			1200		2800					8850					35 300	8760					1 000 000	33 600	1 500 000		870 000	
	Solid (%) WY:96-98			82		89					91					90	50					99	94	100		71	
Columbia River near Beaver Army Terminal	Flux (S) WY:96	16·8	<8	824	403	2860	13	980	300	600	9410	1850	470	25	<84	21 900	5480	120	17	7	1	740 000	18 500	1 300 000	85 700	340 000	370 000
	Flux (D) WY:96			462		560		360			5080					6790	21 700	300				8350	920	6180		540 000	
	Flux (T) WY:96			1285		3410		1330			14 500					28 600	27 000	430				750 000	19 400	1 300 000		880 000	
	Solid (%) WY:96			64		84		73			65					76	20	29				99	95	100		38	
	Flux (S) WY:97	16·5	<8	808	396	2800	13	960	300	600	9240	1820	460	25	<82	21 400	5280	120	16	7	1	730 000	18 100	1 300 000	84 100	330 000	360 000
	Flux (D) WY:97			417		560		400			5180					6980	22 000	320				5580	850	3780		590 000	
	Flux (T) WY:97			1225		3360		1360			14 400					28 400	27 300	440				740 000	19 000	1 300 000		920 000	
	Solid (%) WY:97			66		83		70			64					75	19	27				99	96	100		36	
	Flux (S) WY:98	4·5	<2	222	109	770	4	260	82	160	2540	500	130	7	<23	5900	1450	33	5	2	0·3	200 000	4990	350 000	23 100	90 700	99 800
	Flux (D) WY:98			265		420		300			4190					4630	17 700	230				1930	460	1240		400 000	
	Flux (T) WY:98			487		1190		560			5730					10 500	19 100	260				200 000	5440	350 000		490 000	
	Solid (%) WY:98			46		65		47			38					56	8	13				99	92	100		19	
	Flux (S) WY:96-98	37·8	<19	1854	908	6430	30	2200	680	1360	21 200	4160	1060	57	<189	49 200	12 109	280	38	15	3	1 700 000	41 600	2 900 000	190 000	760 000	830 000
	Flux (D) WY:96-98			1143		1530		1060			14 500					18 400	61 400	860				15 900	2230	11 200		1 500 000	
	Flux (T) WY:96-98			2998		8000		3250			35 600					67 600	73 500	1130				1 700 000	43 900	2 900 000		2 300 000	
	Solid (%) WY:96-98			62		81		67			59					73	16	24				99	95	100		33	

between Warrendale and Beaver Army Terminal after the Willamette River enters the mainstem Columbia River (Table VI).

Sediment-associated TOC and P, which apparently are unaffected by mining-related or other activities upstream of Northport, also steadily increase downstream through the Columbia Basin (Table VI). As with sediment-associated trace element fluxes, sediment-associated TOC and P fluxes seem to be affected by inputs from the Snake and Willamette Rivers. In particular, note the marked increase in TOC between Vernita and Warrendale; this may be related to inflows from the Snake River, but also may result from agricultural activities in the unsampled Yakima, Deschutes, and John Day Basins. Finally, note the significant increase in sediment-associated P fluxes, from 61% to 73%, between the Warrendale and Beaver Army Terminal sites after the Willamette River flows into the mainstem Columbia River (Table VI). This may be related to the higher population densities in the Willamette River Basin, and particularly to the city of Portland, relative to the rest of the Columbia River Basin.

THE RIO GRANDE BASIN

Basic geographic data covering the Rio Grande Basin are provided in Table I and a schematic of the sampling sites is provided in Figure 3. Mean and median chemical concentrations for suspended sediment-associated trace elements from the basin are provided in Table II. Sediment-associated trace element concentrations for the Rio Grande seem typical for fine-grained sediment (Table II) (Horowitz, 1991; Persaud *et al.*, 1993). In fact, there is little chemical difference in sediment-associated trace element concentrations throughout the mainstem Rio Grande (Table II).

During the period of study, a long-term drought occurred in the basin, which has likely skewed the results reported herein relative to the long-term record for the region (Hooper *et al.*, 2001). With the exception of the El Paso and Arroyo Colorado sites, where discharge has been maintained by reservoir releases at the former and pumping at the latter, flows have been less than 50% of the annual average (Table VIII). The effects of the drought on basinwide discharges have been exacerbated by the increased withdrawals needed to meet irrigation requirements. The impact at Brownsville has been dramatic; discharge has decreased by more than 95% (Table VIII).

Annual suspended sediment fluxes for the Rio Grande to the Gulf of Mexico (Rio Grande at Brownsville) for the study period averaged about 9600 tonnes (Table IX). This is only slightly above 1% of the annual average (700 000 tonnes) (Table I), and is the direct result of the severe regional drought, compounded by the local installation of a temporary dam designed to raise water levels to facilitate pumping for irrigation (D. Lurry, USGS, oral communication, 2000).

Table VIII. Comparison of average current and historic discharges for the sampling sites in the Rio Grande Basin

Sample site	Historic discharge[a] ($m^3\ s^{-1}$)	1996 discharge ($m^3\ s^{-1}$)	1997 discharge ($m^3\ s^{-1}$)	1998 discharge ($m^3\ s^{-1}$)
Rio Grande at El Paso	17·4	18·4	18·3	18·1
Rio Grande at Foster Ranch	55·1	24·7[b]	25·5[c]	15·1
Rio Grande at Laredo	97·2	51·0	40·3	73·6[d]
Rio Grande near Brownsville	63·0	2·9	2·8	2·9
Arroyo Colorado at Harlingen	7·0	5·7	7·7	5·9

[a] International Boundary and Water Commission, El Paso, TX.
[b] Skewed by the highly localized effects of tropical depressions during August and September 1996.
[c] Skewed by the highly localized effects of tropical depressions during October 1996 and May–June 1997.
[d] Skewed by the highly localized effects of a tropical depression during August and September 1998.

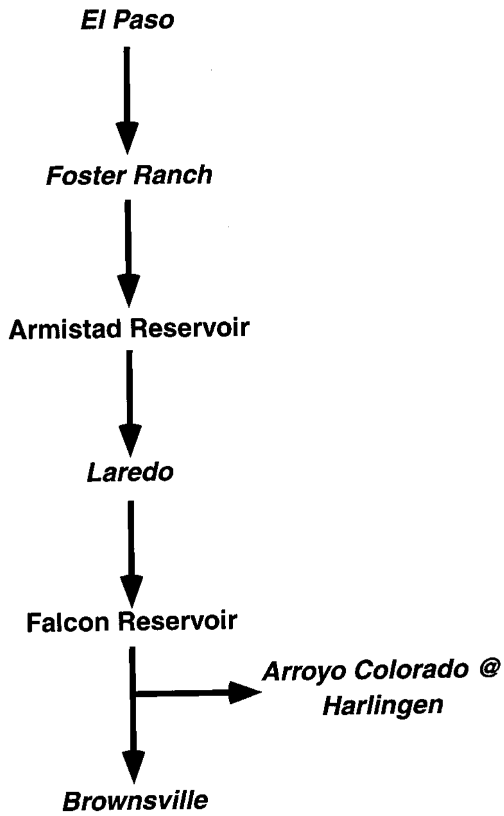

Figure 3. A schematic diagram of the NASQAN Rio Grande Basin; italicized sites represent actual sampling sites (not to scale). The arrows indicate downstream flow direction; note that the Arroyo Colorado diverts flow from the Brownsville site

Despite its size, and as a result of the relatively arid conditions extant, discharge, and hence suspended sediment fluxes in the Rio Grande Basin are both highly localized and extremely 'flashy'. The relatively localized nature of this pattern is compounded by the presence of Amistad and Falcon Reservoirs which temper downstream effects (Figure 3). This conclusion is particularly obvious from an examination of the interannual variations (based on a combination of actual and predicted values) in suspended sediment fluxes for the Foster Ranch and Laredo sites (Table IX). Suspended sediment transport at Foster Ranch for the study period was about 9 Mt. The majority of this material (77%) was transported in 1996 and secondarily, in 1997 (19%). In 1996, two events accounted for 94% of the annual flux; between August 29 and September 8, 1996, and between September 14 and September 27, 1996, 2·4 and 4·1 Mt, respectively, of suspended sediment were transported through the Foster Ranch site as a result of localized tropical depressions. The August–September event occurred as a direct result of the remnants of Hurricane Dolly, which moved inland between New Mexico and Texas, and substantially impacted discharges in the Rio Conchos, which flows into the Rio Grande upstream of Foster Ranch. In 1997 as in 1996, and again as the result of localized tropical depressions, two events accounted for 91% of the annual flux. Between October 8 and October 28, 1996, and between May 10 and June 30, 1997, 0·64 and 0·91 Mt, respectively, of suspended sediment passed the Foster Ranch site. The presence of Amistad Reservoir seems to have prevented the impacts of these events from reaching the next downstream sampling site at Laredo (Table IX).

A similar pattern to that for Foster Ranch also can be observed at the Laredo site (Table IX). Suspended sediment transport at Laredo for the study period totalled about 1·1 Mt. The majority of this material was transported in 1998 during a 4-day period. Between August 25 and August 28, 1998, 0·71 Mt of suspended

Table IX. Summary of estimated Rio Grande Basin fluxes for the 1996, 1997, and 1998 water years (see footnote for Table III)

Location		S.S. (Mt)	Ag (t)	Cu (t)	Pb (t)	Zn (t)	Cd (t)	Cr (t)	Co (t)	Ni (t)	Ba (t)	V (t)	Li (t)	Be (t)	Mo (t)	P (t)	Sr (t)	As (t)	Sb (t)	Se (t)	Hg (t)	Fe (t)	Mn (t)	Al (t)	Ti (t)	TOC (t)	Tot. C (t)
Rio Grande at El Paso	Flux (S) WY:96	260 000	<0·1	5	5	21	0·1	11	2	6	150	12	12	0·4	<1	180	123	1	0·2	0·08	0·008	4980	260	13 900	660	2880	7340
	Flux (D) WY:96			1		3		1		1	41		57		5	22	514	1				4	4	2		3090	
	Flux (T) WY:96			6		23		12		7	190		69			210	637	3				4980	270	13 900		5970	
	Solid (%) WY:96			83		89		91		86	79		17			89	19	47				100	99	100		48	
	Flux (S) WY:97	310 000	<0·2	6	6	24	0·1	13	2	7	180	14	14	0·4	<1·5	220	145	2	0·3	0·09	0·009	5850	310	16 300	770	3390	8620
	Flux (D) WY:97			1		2		1		1	40		55		5	21	490	2				1	4	2		2330	
	Flux (T) WY:97			7		26		14		8	220		69			240	635	3				5850	310	16 300		5710	
	Solid % WY:97			85		93		92		88	0		20			91	23	47				100	99	100		59	
	Flux (S) WY:98	270 000	<0·1	5	5	21	0·1	12	2	6	160	12	12	0·4	<1·4	190	127	1	0·2	0·08	0·008	5140	270	14 300	680	2980	7580
	Flux (D) WY:98			1		1		1		1	41		56		5	21	500	2				3	4	2		1780	
	Flux (T) WY:98			6		23		13		8	200		68			210	628	3				5150	270	14 300		4760	
	Solid (%) WY:98			84		95		91		87	79		18			90	20	40				100	99	100		63	
	Flux (S) WY:96-98	840 000	<0·4	16	17	66	0·3	36	7	20	490	38	38	1	<4·2	590	395	4	0·8	0·25	0·03	16 000	840	44 600	2100	9250	23 500
	Flux (D) WY:96-98			3		6		3		3	120		170		15	64	1504	5				8	12	7		7200	
	Flux (T) WY:96-98			19		72		39		23	610		210			650	1899	9				16 000	850	44 600		16 400	
	Solid (%) WY:96-98			84		92		92		87	80		18			90	21	44				100	99	100		56	
Rio Grande at Foster Ranch	Flux (S) WY:96	6 900 000	<3·4	100	120	610	2	310	55	140	1990	590	280	11	<34	5420	5075	55	7	5	0·3	160 000	3360	420 000	19 200	75 400	280 000
	Flux (D) WY:96			2		2		1		2	82	4	52		6	12	1280	1		1		1	1	2		2340	
	Flux (T) WY:96			100		610		310		150	2070	590	330			5430	6354	56		6		160 000	3360	420 000		77 800	
	Solid (%) WY:96			98		100		100		99	96	99	84			100	80	98		84		100	100	100		97	
	Flux (S) WY:97	1 700 000	<0·9	26	30	160	0·5	79	14	37	510	150	72	3	<8·7	1380	1291	14	2	1	0·09	41 900	860	110 000	4880	19 200	71 500
	Flux (D) WY:97			2		2		2		2	89	5	54		7	13	1336	1		1		1	1	2		2440	
	Flux (T) WY:97			28		160		80		38	600	160	130			1390	2627	15		2		41 900	860	110 000		21 630	
	Solid (%) WY:97			94		99		98		96	85	97	57			99	49	91		55		100	100	100		89	
	Flux (S) WY:98	420 000	<0·2	6	7	37	0·1	19	3	9	120	36	17	0·7	<2·1	330	309	3	0·4	0·3	0·02	10 000	200	25 500	1170	4590	17 100
	Flux (D) WY:98			1		1		1		1	48	4	38		4	8	870	1		1		2	1	1		1180	
	Flux (T) WY:98			7		38		20		10	170	40	55			340	1179	4		1		10 000	200	25 500		5770	
	Solid (%) WY:98			89		98		95		92	72	89	31			98	26	76		35		100	100	100		80	
	Flux (S) WY:96-98	9 000 000	<4·5	140	150	800	3	410	72	190	2620	780	370	14	<45	7130	6675	72	9	6	0·5	220 000	4420	550 000	25 300	99 200	370 000
	Flux (D) WY:96-98			4		4		4		4	220	13	140		17	33	3486	4		2		4	2	4		5960	
	Flux (T) WY:96-98			140		810		410		190	2830	790	510			7160	10 160	76		9		220 000	4420	550 000		110 000	
	Solid (%) WY:96-98			97		99		99		98	92	98	72			100	66	95		72		100	100	100		94	
Rio Grande at Laredo	Flux (S) WY:96	120 000	<0·06	2	2	12	0·05	5	1	2	44	9	4	0·1	<0·6	110	70	1	0·1	0·07	0·007	2690	54	6480	330	2080	5870
	Flux (D) WY:96							4		2	170				11	90	2300					8	8	8		3910	
	Flux (T) WY:96							9		4	220					200	2370					2700	62	6480		5990	
	Solid (%) WY:96							52		47	20					55	3					100	87	100		35	
	Flux (S) WY:97	74 400	<0·04	1	1	7	0·03	3	1	1	27	5	3	0·1	<0·4	66	42	1	0·1	0·04	0·004	1640	33	3940	200	1260	3570
	Flux (D) WY:97							3		2	140				8	70	1790					6	6	6		3050	
	Flux (T) WY:97							6		3	170					140	1830					1650	39	3940		4310	
	Solid (%) WY:97							46		41	16					47	2					99	85	100		29	

(*continued overleaf*)

Table IX. (*continued*)

Location		S.S. (Mt)	Ag (t)	Cu (t)	Pb (t)	Zn (t)	Cd (t)	Cr (t)	Co (t)	Ni (t)	Ba (t)	V (t)	Li (t)	Be (t)	Mo (t)	P (t)	Sr (t)	As (t)	Sb (t)	Se (t)	Hg (t)	Fe (t)	Mn (t)	Al (t)	Ti (t)	TOC (t)	Total C (t)
	Flux (S) WY:98	910 000	<0·5	13	16	91	0·4	34	6	15	330	65	33	1·1	<4·5	810	520	7	1	0·5	0·05	20 000	400	48 100	2450	15 400	43 500
	Flux (D) WY:98							4		3	230				14	530	2660					12	5	9		11 000	
	Flux (T) WY:98							38		19	560					1340	3180					20 000	410	48 100		26 400	
	Solid (%) WY:98							89		83	59					60	16					100	98	100		58	
	Flux (S) WY:96-98	1 100 000	<0·55	15	20	110	0·4	41	8	19	400	79	40	1·3	<5·5	980	630	9	1	0·7	0·07	24 300	490	58 500	2980	18 800	53 000
	Flux (D) WY:96-98							12		7	540				32	680	6760					26	19	23		18 300	
	Flux (T) WY:96-98							52		26	940					1660	7390					24 300	510	58 500		37 100	
	Solid (%) WY:96-98							78		72	43					59	9					100	96	100		51	
Arroyo Colorado at Harlingen	Flux (S) WY:96	53 800	<0·03	1	1	4	0·03	1	0·3	1	11	2	2	0·05	<0·3	40	48	0·3	0·1	0·03	0·01	810	37	1880	86	700	1610
	Flux (D) WY:96			1		1				1	14	3	20		3	39	670	1		0·3		1	1	1		930	
	Flux (T) WY:96			2		5				2	26	6	22			80	720	1		0·3		810	38	1880		1630	
	Solid (%) WY:96			41		80				45	44	42	10			51	7	18		10		100	97	100		43	
	Flux (S) WY:97	72 200	<0·04	1	1	5	0·04	2	0·4	1	15	3	3	0·1	<0·4	54	65	0·4	0·2	0·04	0·01	1080	50	2520	120	940	2160
	Flux (D) WY:97			1		1		1		1	16	6	20		3	71	670	2		0·3		1	1	1		1450	
	Flux (T) WY:97			2		7		2		2	31	9	23			120	740	2		0·3		1080	51	2530		2390	
	Solid (%) WY:97			48		82		78		58	49	33	13			43	9	15		12		100	98	100		39	
	Flux (S) WY:98	55 800	<0·03	1	1	4	0·03	1	0·3	1	12	2	2	0·1	<0·3	42	50	0·3	0·1	0·03	0·01	840	38	1950	89	720	1670
	Flux (D) WY:98			1		1		1		1	14	4	19		3	72	660	1		0·3		2	1	1		900	
	Flux (T) WY:98			1		5		2		1	26	6	22			110	710	2		0·3		840	40	1950		1630	
	Solid (%) WY:98			54		80		74		61	46	39	11			37	7	17		10		100	97	100		44	
	Flux (S) WY:96-98	180 000	<0·09	2	3	14	0·09	5	1	3	38	8	8	0·2	<0·9	140	160	1	0·4	0·1	0·02	2730	120	6360	290	2360	5450
	Flux (D) WY:96-98			3		3		1		2	44	13	58		10	180	2000	5		1		4	4	3		3280	
	Flux (T) WY:96-98			5		17		6		5	82	21	66			320	2160	5		1		2730	130	6360		5640	
	Solid (%) WY:96-98			47		81		76		54	46	37	12			43	8	17		10		100	97	100		42	
Rio Grande near Browns-ville	Flux (S) WY:96	11 600	<0·01	0	0·4	2	0·006	0·5	0·1	0·3	6	1	1	0·02	<0·1	17	9	0·2	0·03	0·01	0·002	320	21	700	32	390	710
	Flux (D) WY:96			0		0·3					9		4		1	4	130	0·4	0·01			0·2	0·1	0·2		480	
	Flux (T) WY:96			0		2					15		5			21	140	1	0·04			320	21	700		870	
	Solid (%) WY:96			58		84					38		13			80	7	29	75			100	99	100		45	
	Flux (S) WY:97	8300	<0·01	0·2	0·3	1	0·004	0·4	0·1	0·2	4	1	0	0·01	<0·1	12	7	0·1	0·02	0·01	0·001	230	15	500	23	280	510
	Flux (D) WY:97			0·1		0·2					9		4		1	7	120	0·4				0·4	0·1	0·2		410	
	Flux (T) WY:97			0·3		1					13		4			19	120	0·5				230	15	500		690	
	Solid (%) WY:97			53		83					30		10			64	5	24				100	99	100		40	
	Flux (S) WY:98	8800	<0·01	0·2	0·3	1	0·004	0·4	0·1	0·2	4	1	0	0·01	<0·1	13	7	0·1	0·02	0·01	0·001	240	16	530	24	300	540
	Flux (D) WY:98			0·1		0·2					10		4		1	16	120	0·4				0·2	0·2	0·2		440	
	Flux (T) WY:98			0·3		1					14		4			29	120	0·5				240	16	530		740	
	Solid (%) WY:98			59		85					30		11			44	6	24				100	99	100		40	
	Flux (S) WY:96-98	28 700	<0·02	1	1	4	0·01	1	0·2	1	14	2	2	0·05	<0·2	41	23	0·4	0·07	0·03	0·005	790	52	1730	79	970	1760
	Flux (D) WY:96-98			0·4		1		0·4		0·5	28		12		2	27	360	1				1	0·4	1		1340	
	Flux (T) WY:96-98			1		5		2		1	42		13			68	380	1				800	52	1730		2310	
	Solid (%) WY:96-98			57		84		73		61	33		11			61	6	26				100	99	100		42	

sediment passed the Laredo site as a result of tropical depression Charlie, which began as a major hurricane in the Gulf of Mexico. The effects of the tropical depression accounted for 79% of the suspended sediment transported during the 1998 water year, as well as 82% of the suspended sediment transported at the site for the three combined water years (Table IX). As at Foster Ranch, the presence of a major impoundment (Falcon Reservoir) seems to have prevented the impact of this event from reaching the next downstream sampling site at Brownsville (Table IX).

As noted for both the Mississippi and Columbia River Basins, the majority of the trace element fluxes in the Rio Grande Basin occur in association with the movement of suspended sediment (Table VI). Considering the extremely flashy and localized nature of suspended sediment fluxes in the Rio Grande Basin, and particularly the presence of both Amistad and Falcon Reservoirs (Figure 3), and unlike the highly regulated Columbia River (see previous section), there do not seem to be any continuous downstream increases in either sediment-associated or total trace element fluxes (Tables VI and IX). Sediment-associated and total trace element fluxes do increase between the El Paso and the Foster Ranch sites (Tables VI and IX; Figure 3). This probably results from the nearly three-fold increase in the drainage area of the basin between these two sites combined with a lack of any intervening impoundments (Figure 3) (Hooper *et al.*, 2001). However, between the Foster Ranch site and the Laredo site, there is a nearly nine-fold decline in both sediment-associated and total trace element fluxes (Tables VI and IX). This occurs despite a nearly four-fold increase in drainage area (Hooper *et al.*, 2001) and a nearly two-fold increase in discharge (Table VIII). Presumably, this is due to deposition in Amistad Reservoir, which is located between the two sites. A similar declining pattern of suspended sediment, as well as sediment-associated and total trace element fluxes occurs between the Laredo and the Brownsville sites (Tables VI and IX). This results from the nearly 20-fold decline in discharge and the nearly 40-fold decline in suspended sediment fluxes between the two (Tables VIII and IX). The decline in discharge and suspended sediment fluxes between Laredo and Brownsville partially results from the diversion of a portion of the flow of the Rio Grande into the Arroyo Colorado. This diversion seems to account for about 15 to 20% of the discharge and suspended sediment fluxes passing through the Laredo site (Table IX). Further declines in discharge probably can be attributed to increased demands for irrigation water as a result of the long-term drought, whereas declines in suspended sediment and sediment-associated trace element fluxes probably can be attributed to sediment deposition in Falcon Reservoir, which lies between Laredo and Brownsville (Figure 3).

THE COLORADO BASIN

Basic geographic data covering the Colorado River Basin are provided in Table I, and a schematic of the sampling sites is provided in Figure 4. As a result of the numerous and closely spaced dams located in the system, and the fact that little or no sediment passes through them, the Colorado River may well be the most regulated of the four NASQAN basins (Figure 4) (Hooper *et al.*, 2001). Mean and median chemical concentrations for suspended sediment-associated trace elements from the basin are provided in Table II. Sediment-associated trace element concentrations for the Colorado Basin seem typical for fine-grained sediment (Table II) (Horowitz, 1991; Persaud *et al.*, 1993). In fact, there is little chemical difference in sediment-associated trace element concentrations throughout the basin (Table II). However, the Zn, V, and TOC concentrations for suspended sediment collected at the Colorado River above Diamond Creek do seem somewhat low relative to the concentrations determined for the other sites (Table II). It should be noted that the Colorado River, probably as a result of local petrology combined with irrigation return flow, is the only NASQAN basin where dissolved Se consistently occurs above detection limits.

Annual suspended sediment fluxes entering Mexico at the Northern International Boundary (NIB) near Andrade, CA, for the study period averaged about 365 000 tonnes (Table X). This is more than 3·5 times above the published annual average (Table I; Meade and Parker, 1985). However, the use of average values is misleading because the discharges for each water year were markedly different; the discharge for 1996 was

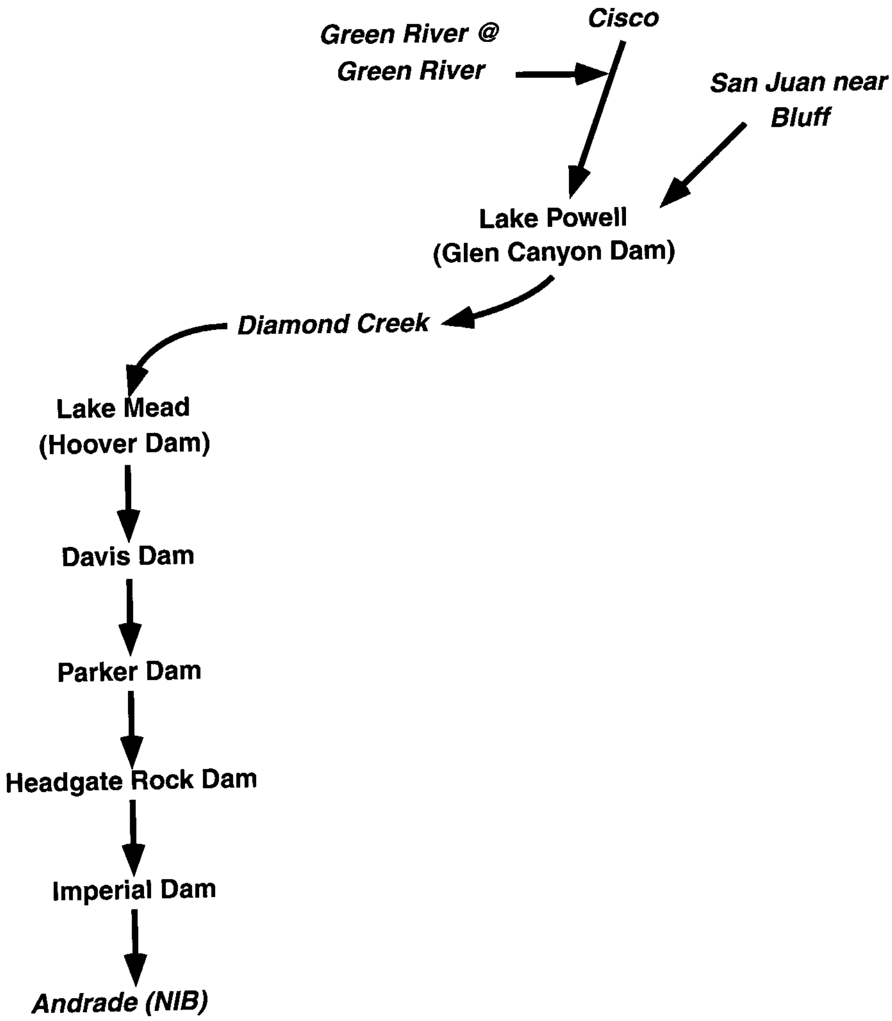

Figure 4. A schematic diagram of the NASQAN Colorado River Basin; italicized sites represent actual sampling sites (not to scale). The arrows indicate downstream flow direction

slightly below average, whereas the discharges for 1997 and 1998 were three and seven times higher than average, respectively (Table X). The differences at NIB are not reflected in the middle (Diamond Creek) or the upper parts (Colorado near Cisco, Green River, and Bluff) of the basin (Figure 4; Table X) and seem to be the direct result of substantial annual increases in both discharge and suspended sediment concentrations. In 1996, discharge and suspended sediment concentration averaged 58 m^3 s^{-1} and 85 mg l^{-1}, respectively, whereas in 1997 they averaged 99 m^3 s^{-1} and 146 mg l^{-1}, respectively, and in 1998 they averaged 143 m^3 s^{-1} and 211 mg l^{-1}, respectively (Horowitz *et al*., 2001; unpublished data). These increased discharges resulted from above average releases from the dams downstream from the Diamond Creek sampling site (Figure 4). The amount of water released in 1997 and 1998, which were surplus water years, was 1·6 and 6·4 times the amount, respectively, released in 1996 (P. Matuska, Bureau of Reclamation, oral communication, 1999). Further, an increase in sediment availability can be inferred as a result of material left in storage upstream of the NIB site, as a result of the 1993 Gila River floods (K. Fagot, Bureau of Reclamation, oral communication, 1999).

Table X. Summary of estimated Colorado Basin fluxes for the 1996, 1997, and 1998 water years (see footnotes for Table III)

Location		S.S. (Mt)	Ag (t)	Cu (t)	Pb (t)	Zn (t)	Cd (t)	Cr (t)	Co (t)	Ni (t)	Ba (t)	V (t)	Li (t)	Be (t)	Mo (t)	P (t)	Sr (t)	As (t)	Sb (t)	Se (t)	Hg (t)	Fe (t)	Mn (t)	Al (t)	Ti (t)	TOC (t)	Tot. C (t)
Colorado River near Cisco	Flux (S) WY:96	4·0	<2·0	76	100	380	3	210	28	110	1620	280	140	5	<20	2440	1600	28	6	7	0·2	75 800	1760	180 000	8380	59 900	120 000
	Flux (D) WY:96			20		18		9		11	360		130		36	150	4180	7		18		88	49	43		31 800	
	Flux (T) WY:96			96		400		220		120	1990		270			2580	5780	35		25		75 900	1810	180 000		91 700	
	Solid (%) WY:96			79		96		96		91	82		51			94	28	79		28		100	97	100		65	
	Flux (S) WY:97	9·8	<4·9	180	250	940	7	520	68	270	4000	680	340	12	<49	5950	3900	68	16	17	0·4	190 000	4290	450 000	20 500	150 000	290 000
	Flux (D) WY:97			19		18		15		13	490		170		39	170	4940	10		19		95	43	54		73 100	
	Flux (T) WY:97			200		950		530		290	4490		510			6120	8840	78		36		190 000	4340	450 000		220 000	
	Solid (%) WY:97			91		98		97		96	89		67			97	44	87		46		100	99	100		67	
	Flux (S) WY:98	4·2	<2·1	80	110	410	3	220	30	120	1740	300	150	5	<21	2580	1690	30	7	7	0·2	80 500	1860	190 000	8900	53 500	130 000
	Flux (D) WY:98			11		10		13		11	410		180		30	100	4540	8		18		30	28	21		84 200	
	Flux (T) WY:98			91		420		240		130	2150		330			2690	6230	37		25		80 500	1890	190 000		150 000	
	Solid (%) WY:98			88		98		94		92	81		45			96	27	80		28		100	99	100		43	
	Flux (S) WY:96-98	18·0	<9·0	340	470	1730	13	950	130	500	7370	1260	630	22	<90	11 000	7190	126	29	31	0·7	340 000	7910	830 000	37 800	270 000	540 000
	Flux (D) WY:96-98			50		45		37		34	1260		480		105	430	13 700	25		55		210	119	118		190 000	
	Flux (T) WY:96-98			390		1770		990		540	8630		1110			11 400	20 800	151		86		340 000	8030	830 000		460 000	
	Solid (%) WY:96-98			87		97		96		94	85		57			96	35	84		36		100	99	100		59	
Green River at Green River	Flux (S) WY:96	5·7	<2·8	97	97	400	2	340	40	160	2900	360	220	7	<29	3870	1710	40	5	3	0·2	110 000	1990	260 000	11 400	57 000	130 000
	Flux (D) WY:96			14		10		7		8	340		120		15	110	3030	6		7		42	7	21		45 800	
	Flux (T) WY:96			110		400		340		170	3240		330			3980	4740	46		10		110 000	2000	260 000		100 000	
	Solid (%) WY:96			88		98		98		95	90		65			97	36	86		29		100	100	100		55	
	Flux (S) WY:97	9·6	<4·8	160	160	680	4	570	67	280	4890	600	360	12	<48	6520	2880	67	9	5	0·3	180 000	3360	440 000	19 200	95 900	210 000
	Flux (D) WY:97			15		12		12		10	400		140		18	140	3520	10		9		43	9	25		66 300	
	Flux (T) WY:97			180		690		580		290	5290		510			6660	6400	78		14		180 000	3360	440 000		160 000	
	Solid (%) WY:97			92		98		98		97	92		72			98	45	86		35		100	100	100		59	

	Flux (S) WY:98	8·7	<4·3	150	150	620	3	510	61	250	4410	540	330	11	<43	5890	2600	61	8	4	0·3	160 000	3030	400 000	17 300	86 600	190 000
	Flux (D) WY:98			13		13		16		10	430		160		19	150	3740	13		9		10	9	19		64 400	
	Flux (T) WY:98			160		630		530		260	4840		490			6030	6340	73		13		160 000	3040	400 000		150 000	
	Solid (%) WY:98			92		98		97		96	91		68			98	41	83		33		100	100	100		57	
	Flux (S) WY:96-98	23·9	<12·0	410	410	1700	10	1410	170	690	12 200	1510	910	31	<120	16 300	7180	168	22	12	0·7	450 000	8380	1 100 000	47 900	240 000	530 000
	Flux (D) WY:96-98			42		35		35		28	1160		420		52	400	10 300	29		25		95	24	66		180 000	
	Flux (T) WY:96-98			450		1740		1450		720	13 400		1330			16 700	17 500	197		37		450 000	8400	1 100 000		420 000	
	Solid (%) WY:96-98			91		98		98		96	91		69			98	41	85		33		100	100	100		58	
San Juan River near Bluff	Flux (S) WY:96	4·2	<2·1	71	100	330	1	180	29	96	2090	210	110	5	<21	1670	1170	21	4	1	0·1	79 300	1710	220 000	9180	33 400	54 300
	Flux (D) WY:96			4		3		2		2	93		27		2	47	920			1		7	2	8		42 700	
	Flux (T) WY:96			75		330		190		98	2180		140			1720	2090			2		79 300	1710	220 000		76 100	
	Solid (%) WY:96			94		99		99		98	96		81			97	56			52		100	100	100		44	
	Flux (S) WY:97	9·8	<4·9	170	240	780	2	430	69	230	4910	490	260	13	<49	3930	2750	49	9	3	0·3	190 000	4030	520 000	21 600	78 600	130 000
	Flux (D) WY:97			6		5		4		2	160		43		3	100	1450			2		8	3	17		59 700	
	Flux (T) WY:97			180		780		440		230	5080		310			4040	4200			5		190 000	4030	520 000		140 000	
	Solid (%) WY:97			97		99		99		99	97		86			97	66			59		100	100	100		57	
	Flux (S) WY:98	7·3	<3·6	120	170	570	1	320	51	170	3630	360	200	9	<36	2910	2040	36	7	2	0·2	140 000	2980	390 000	16 000	58 100	94 500
	Flux (D) WY:98			3		3		3		2	120		37		2	78	1260			2		7	2	9		53 800	
	Flux (T) WY:98			130		580		320		170	3750		230			2980	3290			4		140 000	2980	390 000		110 000	
	Solid (%) WY:98			97		100		99		99	97		84			97	62			56		100	100	100		52	
	Flux (S) WY:96-98	21·3	<10·6	360	510	1680	4	940	150	490	10 600	1060	570	28	<106	8510	5960	106	19	6	0·6	400 000	8720	1 100 000	46 800	170 000	280 000
	Flux (D) WY:96-98			13		11		9		6	370		110		8	230	3620			5		22	6	34		160 000	
	Flux (T) WY:96-98			380		1690		940		500	11 000		680			8740	9580			11		400 000	8730	1 100 000		330 000	
	Solid (%) WY:96-98			96		99		99		99	97		84			97	62			56		100	100	100		52	

(*continued overleaf*)

Table X. (*continued*)

Location		S.S. (Mt)	Ag (t)	Cu (t)	Pb (t)	Zn (t)	Cd (t)	Cr (t)	Co (t)	Ni (t)	Ba (t)	V (t)	Li (t)	Be (t)	Mo (t)	P (t)	Sr (t)	As (t)	Sb (t)	Se (t)	Hg (t)	Fe (t)	Mn (t)	Al (t)	Ti (t)	TOC (t)	Total C (t)
Colorado River above Diamond Creek	Flux (S) WY:96	8·9	<4·0	120	150	360	2	360	53	180	4100	360	200	9	<44	3740	1510	53	10	2	0·2	140 000	2940	390 000	17 900	44 500	160 000
	Flux (D) WY:96			120		16		24		19	1340		420		59·9	1640	11 500	25		23		22	8	98		2580	
	Flux (T) WY:96			240		380		380		200	5440		620			5380	13 000	78		25		140 000	2950	390 000		47 100	
	Solid (%) WY:96			50		95		95		90	75		32			70	12	68		7		100	100	100		94	
	Flux (S) WY:97	10·5	<5·3	140	180	430	2	420	63	210	4840	420	240	11	<53	4420	1790	53	12	2	0·2	170 000	3470	460 000	21 100	52 600	190 000
	Flux (D) WY:97			56		24		30		24	1620		500		67·8	2980	12 900	27		30		86	30	58		10 700	
	Flux (T) WY:97			200		450		450		230	6460		740			7400	14 700	80		32		170 000	3500	460 000		63 300	
	Solid (%) WY:97			70		96		93		91	75		32			60	12	66		6		100	99	100		83	
	Flux (S) WY:98	10·5	<5·2	140	180	430	2	420	63	210	4820	420	240	10	<52	4400	1780	53	12	2	0·2	170 000	3460	460 000	21 000	52 400	190 000
	Flux (D) WY:98			27		26		28		23	1580		480		60·5	2550	11 300	27		29		74·0	59	28		20 100	
	Flux (T) WY:98			170		460		450		230	6400		720			6950	13 100	80		31		170 000	3520	460 000		72 500	
	Solid (%) WY:98			82		93		93		91	75		33			63	14	66		7		100	98	100		72	
	Flux (S) WY:96-98	29·9	<14·2	390	510	1230	6	1200	180	600	13 800	1200	690	30	<149	12 600	5080	158	33	6	0·6	480 000	9870	1 300 000	59 800	150 000	540 000
	Flux (D) WY:96-98			200		66		82		65	4550		1400		188	7170	35 700	79		82		181	87	184		33 400	
	Flux (T) WY:96-98			590		1300		1280		660	18 400		2090			19 800	40 800	238		88		480 000	9960	1 300 000		180 000	
	Solid (%) WY:96-98			66		95		94		91	75		33			64	12	67		7		100	99	100		83	
Colorado River at NIB	Flux (S) WY:96	0·1	<0·1	3	2	9	0·03	10	1	6	79	8	5	0·2	<0·9	100	79	1	0·3	0	0·01	3280	160	6740	470	1560	3110
	Flux (D) WY:96					13		4		3	200		110		13	17	2300	5		5		3	17	10		4760	
	Flux (T) WY:96					22		14		9	280		120			120	2380	6		5		3290	170	6750		6310	
	Solid (%) WY:96					41		72		69	29		4			86	3	18		3		100	90	100		25	
	Flux (S) WY:97	0·3	<0·3	10	9	34	0·1	38	5	23	300	32	18	1	<3·2	390	300	4	1	0	0·03	12 200	580	25 100	1740	5800	11 600
	Flux (D) WY:97					9		7		5	330		170		20	36	3580	7		7		5	22	17		8100	
	Flux (T) WY:97					43		45		27	620		180			420	3870	11		7		12 200	600	25 100		13 900	
	Solid (%) WY:97					79		85		82	47		10			91	8	36		6		100	96	100		42	

Flux (S) WY:98	0·7	<0·7	22	21	77	0·3	87	10	52	680	72	41	1	<7·4	890	680	9	2	1	0·1	28 100	1330	57 600	3990	13 300	26 600
Flux (D) WY:98					6		10		6	450		220		26	28	4910	9		9		21	28	24		11 600	
Flux (T) WY:98					83		97		58	1130		260			910	5590	18		10		28 100	1360	57 600		24 800	
Solid (%) WY:98					93		90		89	60		16			97	12	49		11		100	98	100		54	
Flux (S) WY:96-98	1·1	<1·1	34	32	119	0·5	140	16	80	1060	110	64	2	<11·4	1380	1060	14	3	2	0·1	43 600	2060	89 500	6200	20 700	41 300
Flux (D) WY:96-98					28		20		14	980		500		59	81	10 800	21		20		28	67	52		24 400	
Flux (T) WY:96-98					147		160		94	2030		560			1460	11 800	34		22		43 600	2130	89 600		45 000	
Solid (%) WY:96-98					81		87		85	52		11			94	9	40		7		100	97	100		46	

In comparison with historical data, the 1996, 1997, and 1998 suspended sediment fluxes in the upper part of the Colorado Basin (Cisco, Green River, and Bluff) were about 30% below average, but within the range of reported variability (Tables I and X). The 3-year average for Cisco was 6·0 Mt (compared to the historical average of $8{\cdot}0 \pm 4{\cdot}9$), whereas the average for Green River was 8·0 Mt (historical average $= 8{\cdot}8 \pm 4{\cdot}4$), and that for Bluff was 7·0 Mt (historical average $= 15{\cdot}4 \pm 7{\cdot}8$) (USGS Suspended Sediment Database, 1999). The sum of the fluxes for these three upper basin sites indicates that the Green River supplied 39%, the San Juan River supplied 15%, and the upper Colorado River supplied 46% of the nearly 63 Mt of sediment transported into Lake Powell (behind Glen Canyon Dam, Figure 4). Although the Colorado at Lees Ferry was not a NASQAN sediment chemical site, use of a limited number of suspended sediment concentration determinations, in conjunction with LOADEST (Crawford, 1996; Horowitz *et al*., 2001), indicates that only a limited amount of sediment (about 360 000 tonnes) passed through this site during 1996, 1997, and 1998 (B. Aulenbach, USGS, unpublished data). Hence, at least 63 Mt (about 21 Mt year^{-1}) were deposited in Lake Powell during the study period. Based on the 1986 Lake Powell survey, 21 Mt year^{-1} is the right order of magnitude, but is about 50% too low because the survey indicated that during its 23-year history the impoundment had lost about 3·2% of its capacity due to the deposition of about 1000 Mt of sediment (Ferrari, 1988). This translates to an annual deposition rate of between 45 and 52 Mt year^{-1}. This difference probably results from three factors: (1) estimation errors in both the sediment flux calculations and in the survey; (2) the below average fluxes for the Cisco, Green River, and Bluff sites (−30%, see above); and (3) the lack of complete assessment of other upper Colorado Basin sources of suspended sediment. An examination of historical data for the Lees Ferry site, prior to the installation of Glen Canyon Dam, indicates that on average, the current three upper Colorado Basin sampling sites account for only about 35% of the suspended sediment in the upper basin (Iorns *et al*., 1964). If the estimates for the three sites are low by 30%, and only accounted for 35% of the suspended sediment in the upper Colorado Basin, then the actual figure should be about 45 Mt year^{-1}. This is quite close to the lower limit calculated from the 1986 Lake Powell survey (Ferrari, 1988).

During 1996, 1997, and 1998, nearly 30 Mt of suspended sediment was transported through the Diamond Creek site (Figure 4; Table X). Because only about 360 000 tonnes of suspended sediment passed through Glen Canyon Dam, the Diamond Creek sediment must represent 'new' material derived from outside either the upper Colorado Basin and/or Lake Powell. The major sources for the suspended sediment load measured at the Diamond Creek site are probably the Little Colorado, Paria, Kanab, and Havasu Rivers (Andrews, 1991).

The estimated fluxes at NIB indicate that the nearly 30 Mt of suspended sediment passing through the Diamond Creek site was reduced to about 1·1 Mt by the time the Colorado River reaches the US/Mexico border. Presumably, the majority of this material was retained in Lake Mead, as well as the various other impoundments located downstream of the Diamond Creek site (Figure 4); albeit a limited amount of sediment probably was deposited along the river itself. Hence, during the study period there was a >98% net loss of suspended sediment between the upper Colorado River and its mouth. If, as surmised, the majority of suspended sediment entering Lake Powell from the upper Colorado Basin is retained there, then the >98% net loss for the entire basin amounts to at least 95 Mt.

As in the other NASQAN basins, the majority of the Cu, Zn, Cr, Ni, Ba, As, Sb, Fe, Mn, Al, P, and TOC is transported in association with suspended sediment, whereas the majority of the Li and Sr is transported by the dissolved phase (Table VI). As indicated above, the Colorado is the only NASQAN basin where dissolved Se was consistently detected; note that the majority of the Se is transported in association with the dissolved phase (Table VI). Not surprisingly, and considering the downstream pattern of declining suspended sediment fluxes, there is a steady decrease in sediment-associated trace element transport downstream from the upper Colorado River to its mouth (Table VI). However, the patterns for P and TOC do not follow those for the trace elements. Sediment-associated P transport declines from the upper to the middle part of the basin, but increases to upper basin levels downstream of the Diamond Creek site (Table VI). In contrast solid phase-associated TOC contributions increase between the upper and middle sections of the basin, and then decline between the middle and lower sections of the basin (Table VI).

Trace element fluxes for the Colorado Basin are presented in Table X. Considering that the majority of trace element transport in the basin occurs in association with the solid phase, that there is a >98% loss of suspended sediment between the upper and lower sections of the basin, and that there are only small differences in sediment-associated trace element concentrations throughout the basin (Table II), it is not surprising that the resultant fluxes display very marked downstream reductions (Table X). The fluxes for Cu, Pb, Cd, Co, Ni, Hg, Fe, Al, Ti, TOC, and total C decline by two orders of magnitude, whereas those for Zn, Cr, Ba, V, Li, Be, P, Sr, As, Sb, Se, and Mn decline by one order of magnitude between the upper Colorado Basin and the NIB site (Table X). Note that the declines in all the fluxes are greater between the middle and lower sections than between the upper and middle sections of the basin (Table X). Here again, this pattern mirrors and seems to be in response to that for suspended sediment.

CONCLUSIONS

(1) In the four NASQAN basins, the majority (typically $\geq$75%) of Cu, Zn, Cr, Ni, Ba, P, As, Fe, Mn, and Al are transported in association with suspended sediment; in contrast, Sr fluxes seem to be dominated by the dissolved fraction, whereas the transport of Li and TOC appears to be divided about equally between both phases.

(2) Average dissolved trace element concentrations determined during the first 3 years of the NASQAN programme are markedly lower than those reported during the original NASQAN programme; this seems to be due to the use of 'clean' sampling, processing, and analytical techniques, rather than to an improvement in water quality.

(3) The majority of suspended sediment-associated trace element concentrations determined under the NASQAN programme do not seem to be elevated and are typical for fine-grained sediment. However, there are a limited number of exceptions, including: elevated Zn levels in the Ohio River; elevated Hg levels in the Tennessee River; and elevated Ag, Cu, Pb, Zn, Cd, Cr, Co, Ba, Mo, Sb, Hg, and Fe levels in the upper Columbia River.

(4) Annual suspended sediment fluxes in the Mississippi River Basin seem somewhat low relative to previously published averages, but are well within the range of natural variability. Downstream sediment-associated trace element, P, and TOC fluxes increase in the Mississippi River; the increases are markedly greater in the upper part of the basin (20-fold) than in the middle and lower parts of the basin (1·5-fold).

(5) Suspended sediment fluxes at the mouth of the Columbia River averaged about 25% above the annual average, predominantly as a result of record discharges from the Willamette River in 1996 and 1997. Despite the presence of numerous dams and impoundments, total net suspended sediment fluxes display a fairly simple downstream additive pattern. With the exception of the upper part of the Columbia River Basin, where the results are heavily skewed by mining-related impacts, there is a steady downstream increase in sediment-associated trace element, P, and TOC fluxes.

(6) Annual suspended sediment fluxes at the mouth of the Rio Grande averaged only slightly above 1% of the published annual value, and were the direct result of a severe and ongoing regional drought. Despite its size, and as a result of the relatively arid conditions, as well as the presence of Amistad and Falcon Reservoirs, responses to local tropical depressions moving inland from the Gulf of Mexico can be extreme. Hence, suspended sediment fluxes were both highly localized and extremely 'flashy'. As a result, there were no consistent downstream trends in suspended sediment-associated or total trace element fluxes in the Rio Grande Basin.

(7) Annual suspended sediment fluxes near the mouth of the Colorado River at the US/Mexico border averaged about 3·5 times higher than the published average, due to a surplus of water in the lower Colorado

River Basin in 1997 and 1998. Suspended sediment fluxes markedly decrease downstream in the basin; the majority (>99%) of suspended sediment derived from the upper Colorado River Basin is retained in Lake Powell, whereas the majority of suspended sediment derived from the middle part of the basin is retained in Lake Mead and in the other impoundments in the lower part of the basin. As a result, suspended sediment-associated, as well as total trace element fluxes also decrease downstream; differences between the upper and lower Colorado River Basin typically exceed one order of magnitude, but can be as large as two orders of magnitude.

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