

Heavy Metal Distribution in Sediments of Krishna River Basin, India

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ABSTRACT / Suspended and bed sediments collected from the entire region of the Krishna River and its major tributaries

were analyzed for heavy metals (V, Cr, Mn, Fe, Co, Ni, Cu, Zn, and Pb) by the thin-film energy dispersive x-ray fluorescence technique. There is considerable variation in the concentration of elements towards downstream, which may be due to the variation in the subbasin geology and various degrees of human impact. Suspended particles are enriched in heavy metals throughout the basin relative to bed sediments. The heavy metals are enriched in coarse size fractions (10–90 μm) throughout the Krishna River except its tributary Bhima, where finer fractions (2 μm) dominate. Transition elements correlate very well with each other. There is a striking similarity between the bed sediments of Krishna River and the Indian average. When the annual heavy metal flux carried by the Krishna River was estimated, and viewed in relation to the other major riverine transport, the Krishna is seen to be a minor contributor of heavy metals to the Bay of Bengal.

Introduction

The fate of heavy metals in the aquatic environment is of extreme importance because of their impact on the ecosystem. The metals in such an environment can be accommodated in three basic reservoirs: water, sediment, and biota. Sediments are usually regarded as the ultimate sink for heavy metals discharged into the environment (Gibbs 1973). In a river, suspended sediments contain significantly higher levels of heavy metals than the dissolved phase. For example, an average world river contains 40 and 8.2 $\mu\text{g/l}$ of Fe and Mn, respectively, in the dissolved phase and 48,000 and 1,050 $\mu\text{g/g}$, respectively, as particulates (Martin and Whitfield 1981). Thus a large part of the anthropogenic discharge of heavy metals into the environment becomes part of the suspended matter in rivers, which acts as an efficient scavenger for these metals. Martin and Meybeck (1979) estimated the average river particulate matter composition on a global scale, based on analysis of 40 elements in the Amazon, Congo, Ganges, Magdalens, Mekong, Parana, and Orinoco rivers and compilation of published data on 13 other major world rivers. These 20 major rivers represent 25% of the world drainage area and 15% of the world rivers sediment discharge. However, the present understanding of sediment chemistry is limited because of restricted information available on the large sediment-carrying rivers of Asia. Asian rivers contribute about 50% of the global sediment input to

world oceans (Milliman and Meade 1983). Therefore, the concentrations of heavy metals in the Asian river sediments assume importance in their global budget. Recently, Milliman and others (1987) discussed the human effects on sediment discharge by Asian rivers with Yellow river as an example. Subramanian and others (1987) reported on the heavy metals distribution in the sediments of the Ganges and Brahmaputra rivers. In this report, we present the various factors regulating the heavy metal composition of suspended and bed sediments from the entire Krishna basin, which is the fifth largest river (in terms of catchment area) in the Indian subcontinent.

The Krishna River drains mineralized areas upstream, while it is extensively used for agriculture downstream. It has a drainage area of 258,945 km^2 , covering a distance of 1,400 km from Mahabaleshwar in the western ghats to the river mouth at the Bay of Bengal. Bhima, Tungabhadra, Gataprabha, and Malaprabha are its major tributaries. The river flows through several big cities, namely Raichur, Kurnool, Srisailem and Vijayawada. From the data shown in Table 1, it is evident that Krishna River is one of the most intensively utilized rivers in India, and it may be polluted as a result of various degrees of human impact. The river carries very little sediment load (<4 million t/yr) (Ramesh and Subramanian, 1988) perhaps because of the predominance of precambrian hard rocks, which cover nearly 80% of the basin area (Fig. 1).

Table 1. Water, land, and population data of Krishna River basin^a

Water resources (million cubic meters)					Population			Land	
Potential		Utilized			Total (millions)	Density (km ²)	% rural population	Culturable (1000 ha)	Net irrigated (1000 ha)
Surface runoff	Ground water	Surface runoff	Ground water	Total					
62,784	9,628	52,437	6,513	58,950	38.50	149	80.9	20,299	1,819

^aAfter Chaturvedi 1973.

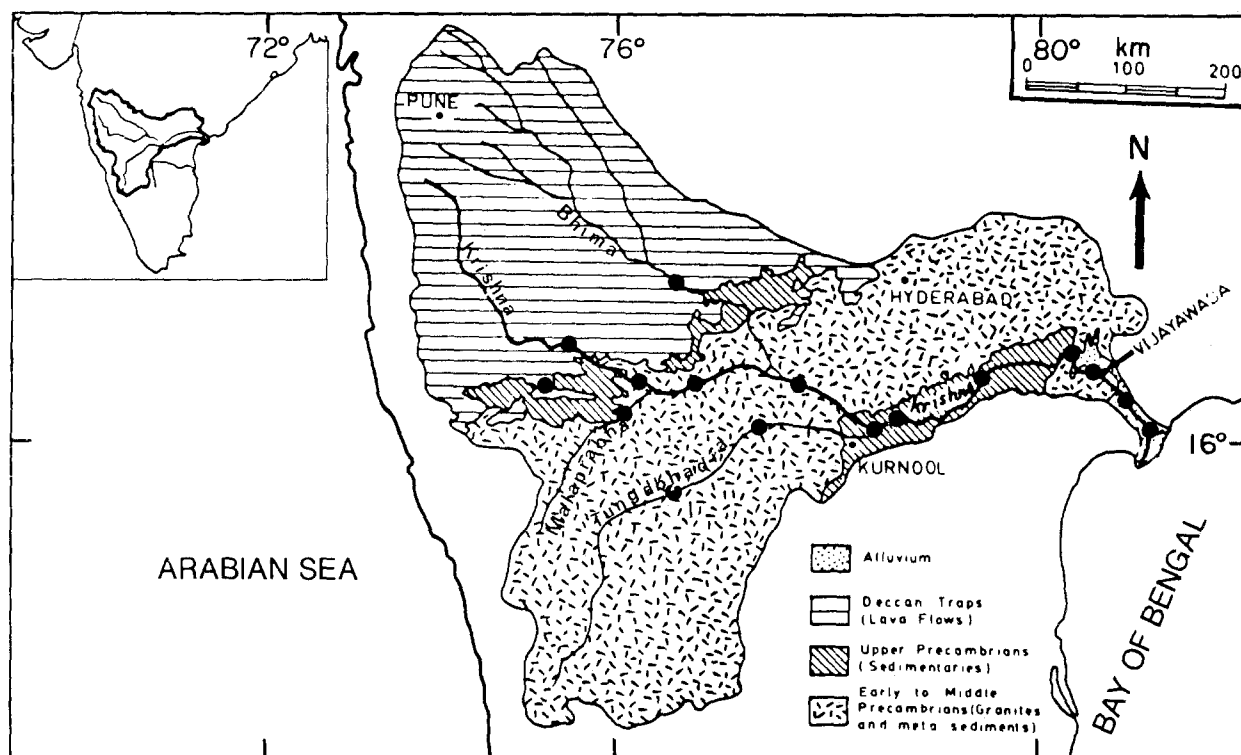


Figure 1. Geological map of the Krishna River basin with sampling locations.

Materials and Methods

Figure 1 shows the geology of the bedrock, general flow direction, and location of sampling sites. During August 1984, 1-l water samples were collected in wide-mouth polyethylene bottles at 16 locations along the river as well as its tributaries. The locations were chosen so as to represent all the regions of the river basin, major urban areas, dams, etc. Suspended sediments were separated by filtration of water through 0.45- μm pore-size membrane filters. In addition, 5 l of water samples were collected in few locations and then separated into clay (2 μm), silt (20 μm), and sand (90

μm) fractions by the conventional pipet method (Guy 1977). Freshly deposited bed sediments from wet portions near the river banks were likewise collected using a small, stainless-steel pipe dredge at 14 locations (Fig. 1) including six samples from the tributaries.

Chemical analysis was performed by the thin-film energy dispersive x-ray fluorescence (XRF) technique. The suspended sediment filters were fitted onto Teflon rings, which were attached to the XRF unit, while the bed sediments, after grinding and suspension, were loaded onto a thin Mylar film and mounted likewise for XRF measurements. Details of this method are presented elsewhere (Van Grieken and

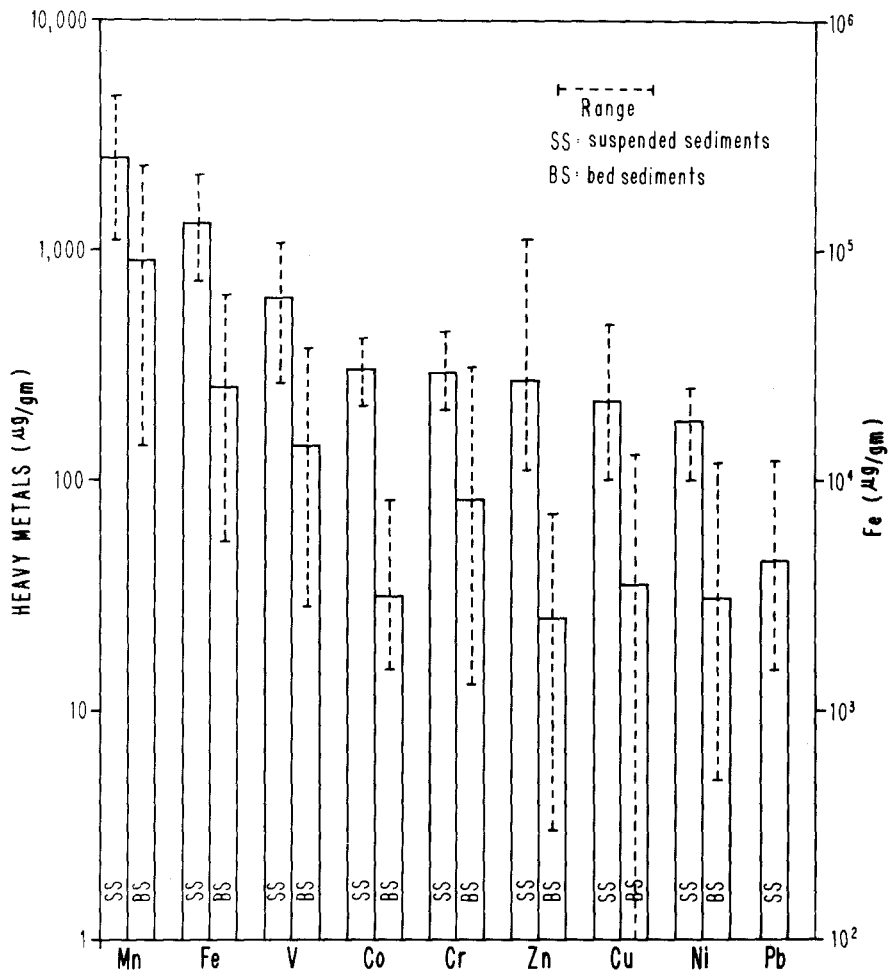


Figure 2. Range and mean concentration of heavy metals in the suspended and bed sediments of the Krishna River basin.

others 1979). The accuracy was checked by analysis of Soil-5 from the International Atomic Energy Agency and BCR-1 from the U.S. Geological Survey. The XRF unit consisted of an HV generator, a Kevex 0810 system with tungsten-x-ray tube, a set of interchangeable secondary fluorescers and filters, a 16-position automatic sampler changer, a 30-mm² Si(Li) detector connected to a multichannel analyzer, and a magnetic tape recorder. The final computer program took into account the various matrix effects (Van Dyck and Van Grieken 1980).

Results and Discussion

Heavy Metals in Suspended and Bed Sediments

The heavy metal contents of the suspended and bed sediments in Krishna River are presented graphi-

cally in Figure 2. Suspended sediment concentrations vary within the basin by a factor of 2–4 for all the elements except Pb and Zn, which vary by a factor of 6 and 11, respectively. On the other hand, the concentration of heavy metals for the bed sediments shows wide variation within the basin. For example, the enrichment factor for Fe, Mn, and Cr is approximately 12, 17, and 24, respectively. Figure 3 shows the downstream profile in the heavy metal content for the suspended sediments (3A) and bed sediments (3B) for the Krishna basin. The points of deflection in the downstream profile are not uniform for both suspended and bed sediments. The variation in concentration towards downstream may be due to the (1) change in relative contribution of sediments draining through different geological formations, (2) size differentiation (sorting) during sediment transport processes, and (3) human influence. Krishna drains very densely popu-

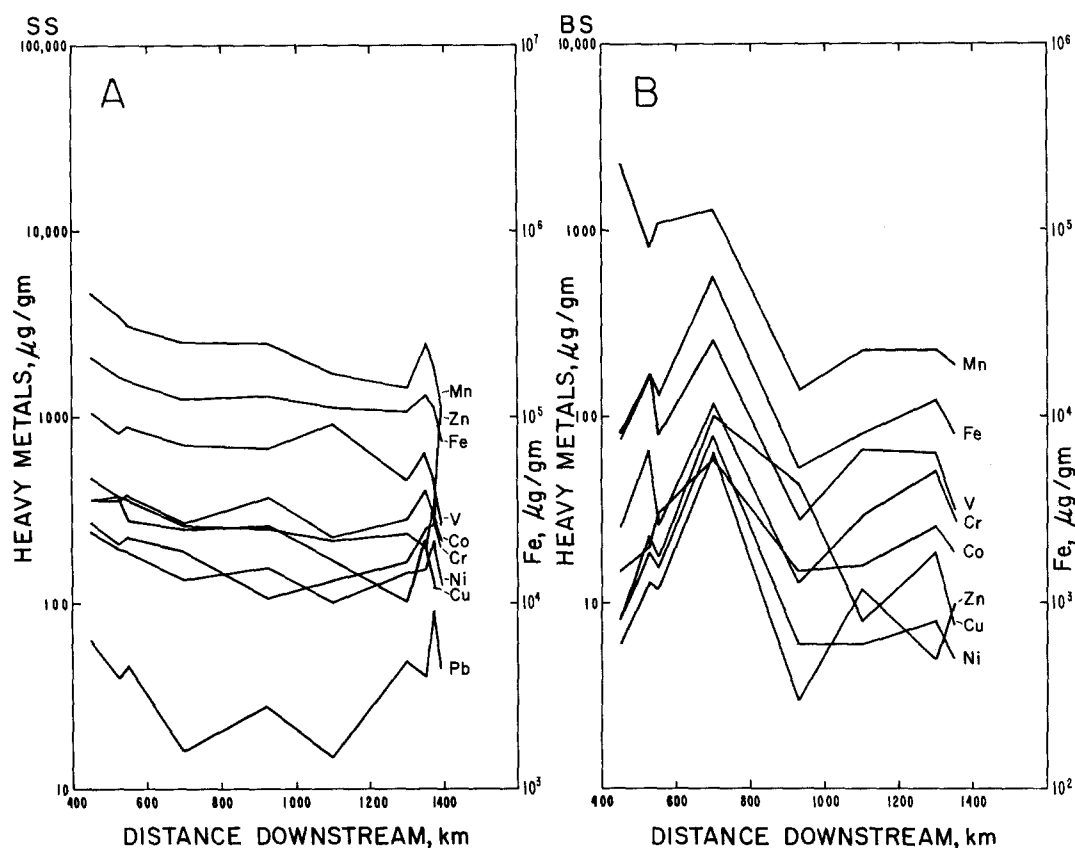


Figure 3. Downstream variation in the concentration of Fe, Mn, Cr, Co, Ni, Cu, Zn, V, and Pb in the suspended (A) and bed sediments (B) of the Krishna River basin.

lated (Table 1) and highly industrial regions. For example, Bhadravathi, known for its steel plant, and Hospet, known for its iron ore deposits, are located on the banks of the River Tungabhadra, a major tributary of Krishna. Hence, the variations of heavy metals do not reflect the true natural geochemical background. There is a sudden depletion in heavy metal concentration for both bed and suspended sediment particles at the river mouth where freshwater sediment enters the estuarine environment (Cl concentration of 81 ppm at Nagayalanka) (Ramesh 1985). Similar observations have been made by Muller and Forstner (1975) for Rhine River sediments. When freshwater mixes with receiving marine waters in estuaries, alkali and alkaline earth cations can actively compete with metal cations sorbed on particle surfaces, and hence the decline of metal concentration in sediment particles.

Figures 2 and 3 illustrate that, for any given location, concentrations of all the heavy metals considered are significantly higher in suspended sediments than in the bed sediments. For example, Fe and Mn are approximately enriched by a factor of 25 and 18 in

suspended sediments when compared to bed sediments, at locations Kolhar and Kurnool, respectively. The suspended sediments are finer and richer in multiple hydroxide coatings (Forstner and Wittmann 1981), organics, and trace metal scavenging clays. In addition, the hydraulic conditions, which influence the movement of bed and suspended material, are different. Hence particulates of most rivers show heavy metal enrichment relative to the bed sediments. For example, Subramanian and others (1987) reported that suspended sediments are 5–10 times richer than the bed sediments in the Ganges and Brahmaputra rivers.

Variation of Heavy Metals with Sediment Particle Size

Both particle size and mineralogy control the elemental composition of riverborne sediments. Quartz and feldspars are normally distributed in a wide size spectrum, whereas clay minerals are dominant in the finer size ranges. The finer fractions also contain higher concentration of trace metals and organic

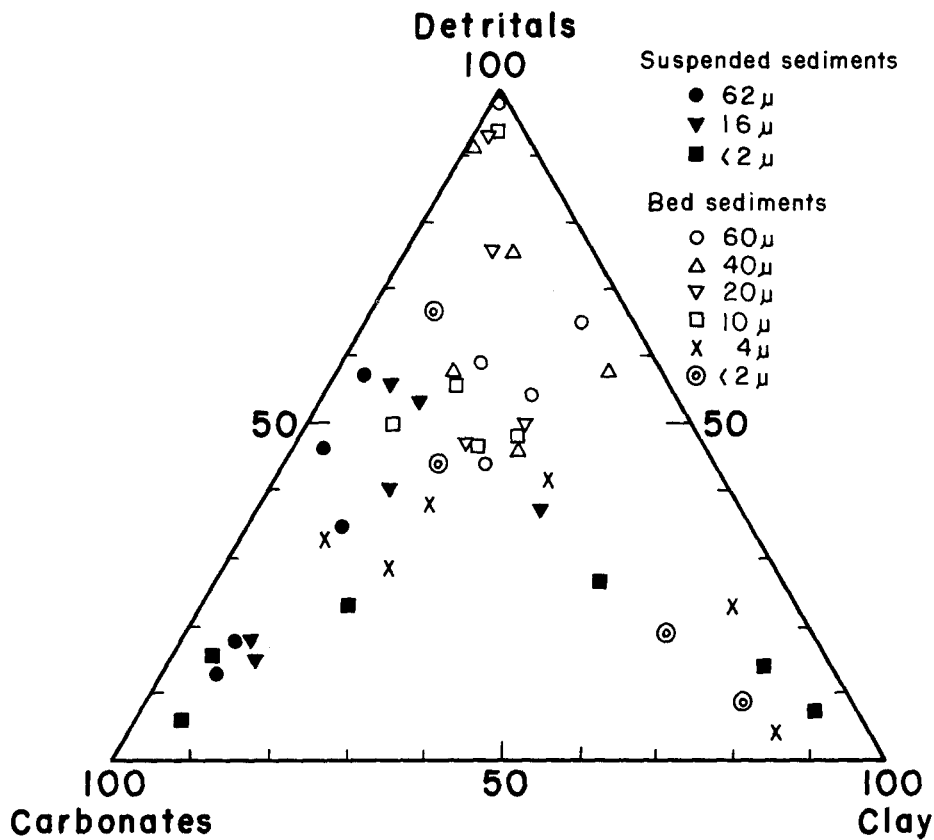


Figure 4. Variation of mineralogy with size in the suspended and bed sediments of the Krishna River basin. Data from Ramesh (1985).

Table 2. Variation of metal concentration ($\mu\text{g/g}$) with sediment particle size in Krishna River basin and its tributaries

	Site ^a														
	Kolhar			Sangam			Vijayawada			TB-Mantralayam			B-Yadgir		
	Clay	Silt	Sand	Clay	Silt	Sand	Clay	Silt	Sand	Clay	Silt	Sand	Clay	Silt	Sand
Fe	30,000	141,400	135,400	51,600	123,400	127,100	26,200	88,100	101,300	29,300	94,300	101,300	88,700	54,300	53,500
Mn	424	3,186	3,036	891	2,717	2,787	267	1,234	1,365	359	2,200	2,371	1,337	987	960
Cr	56	281	249	166	292	263	78	210	301	111	366	346	190	63	71
Co	48	277	226	110	236	241	77	186	262	64	251	235	154	115	129
Ni	34	141	134	64	137	137	34	133	148	53	162	184	90	59	56
Cu	71	329	324	107	266	281	30	107	140	38	107	112	170	121	123
V	93	713	664	232	645	618	72	426	561	137	433	436	408	295	277
Zn	51	203	194	74	171	179	70	225	275	100	207	233	170	85	78
Pb	15	14	9	11	12	10	19	31	41	18	88	109	51	5	8

^aTB—Tungabhadra River; B—Bhima River.

matter. Figure 4 shows the mineral variations with size in the bed and suspended sediments of the Krishna River. Table 2 summarizes the chemical composition of heavy metals in clay, silt, and sand size fractions. In general, contaminated sediments show enrichment of heavy metals in the finer fractions and several authors

(Forstner 1977; Helmke and others 1977) have suggested that the degree of man-made contamination can be properly estimated by examining the pelitic fractions. Figures 5A and 5B indicate the clear dependence of heavy metal content on particle size. From Table 2 and Figures 4 and 5 it is evident that fine frac-

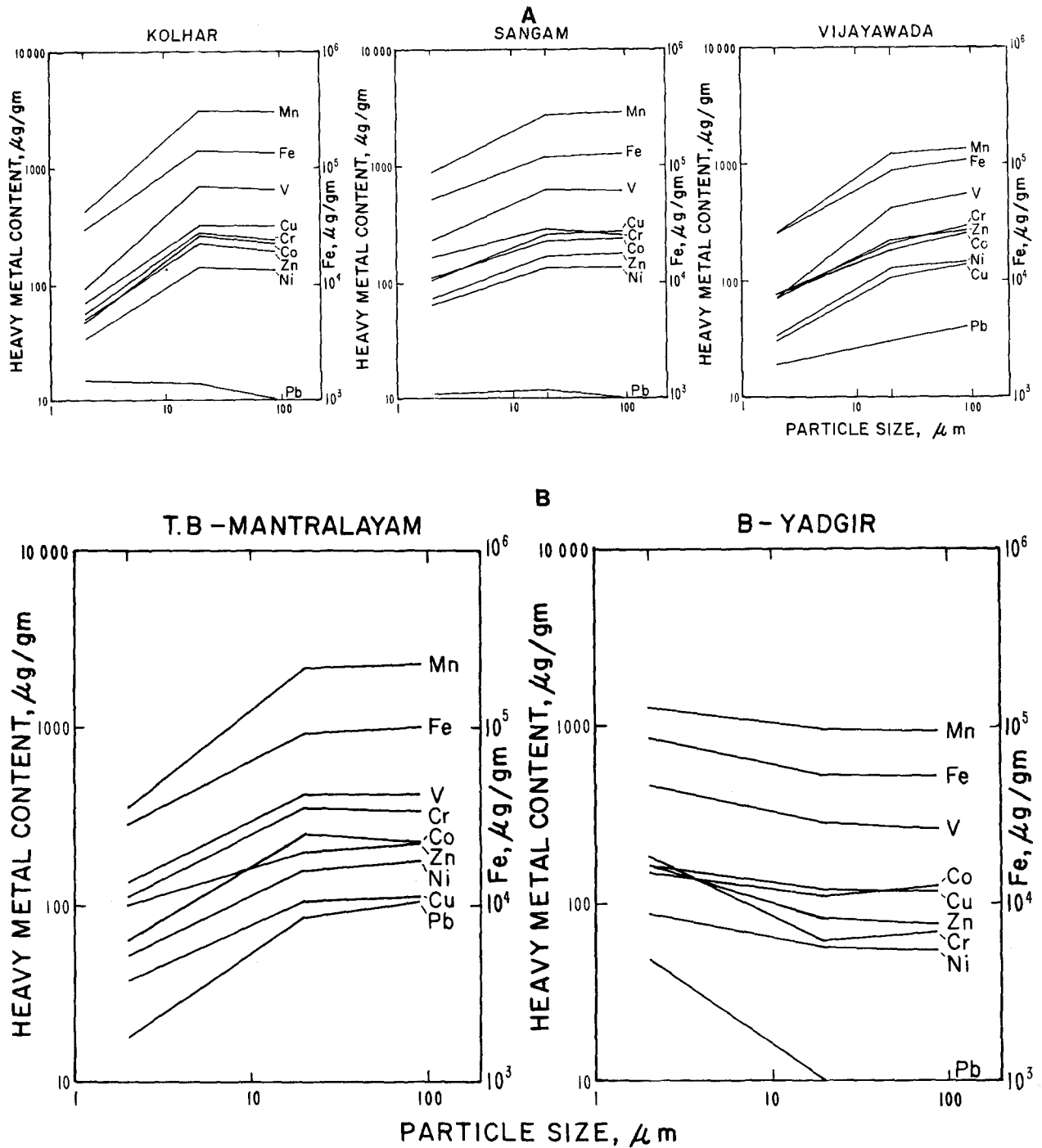


Figure 5A. Variation of metal concentration with particle size in the Krishna River basin. **B.** Variation of metal concentration with particle size in the major tributaries of the Krishna. TB—Tungabhadra; B—Bhima.

tions of Krishna River sediments are neither dominated by clay nor enriched in heavy metals (except Bhima). However, the ratio of concentration of Fe, Pb, and Zn (Table 3) indicates enrichment in finer fractions.

In some cases, mining and smelting water increase the metal concentrations in the coarser size fractions relatively more than in the finer size fractions. For example, Bradley (1982) reported enrichment of heavy metal content in some coarse sediments near mining

Table 3. Size–chemistry relationship for some heavy metals in Krishna River

Location/size	Fe/Mn	Pb/Zn	Pb/Cu	Zn/Cu
Kolhar				
Clay	71	0.29	0.21	0.72
Silt	44	0.07	0.04	0.62
Sand	45	0.05	0.03	0.60
Sangam				
Clay	58	0.15	0.10	0.69
Silt	45	0.07	0.05	0.64
Sand	46	0.06	0.04	0.64
Vijayawada				
Clay	98	0.27	0.63	2.30
Silt	71	0.14	0.29	2.10
Sand	74	0.15	0.29	2.00
Mantralayam				
Clay	82	0.18	0.47	2.6
Silt	43	0.43	0.82	1.9
Sand	43	0.46	0.97	2.1
Yadgir				
Clay	66	0.30	0.30	1.0
Silt	55	0.06	0.04	0.7
Sand	56	0.10	0.07	0.6

areas. However, in the present study no attempt has been made to study the contamination of heavy metals quantitatively. At midstream (Sangam) and downstream (Vijayawada) of Krishna as well as of Tungabhadra, the heavy metals are enriched in the sand size fraction, whereas in the upstream region (Kolhar) the heavy metals are dominated in silt size (Table 2, Fig. 5). On the other hand, for Bhima, one of the major tributaries of Krishna, it is seen that finer fractions are preferentially enriched in heavy metals. Mineralogical changes in river sediments reflect particle size control. For example, Bhima, draining through the Deccan Trap (Fig. 1) carries sediments containing high-exchange-capacity clays such as Montmorillonite. Krishna and Tungabhadra mostly flow through granites and other hard rocks (Fig. 1) essentially constituting detritals (quartz + feldspars) and carbonates even in finer fractions. Thus size chemistry, mineralogy, and pollution input together control the heavy metal composition of the Krishna River sediments.

Using the SAS (Statistical Analysis System) computer software system, Spearman correlation coefficients were computed for some of the heavy metals. Table 4 shows the interelemental relationship between pairs of elements in the combined suspended and bed sediments (except Pb) for Krishna River. In general, all the elements show good correlation with each other. For example, Fe and Mn show excellent correlation with all the elements studied except Zn and Pb.

Although the suspended sediments are richer in heavy metals than the bed sediments, the good correlation in both types indicates that sediments are the ultimate sink for heavy metals in Krishna River sediments, as speculated by several authors (Forstner and Wittmann 1981; Gibbs 1977). These elements are likely to be present together in the organic matter or multiple hydroxide coatings.

Flux of Heavy Metals

Average values for heavy metals in suspended and bed sediments of Krishna River have been computed and are presented in Table 5. Also shown are the published values from major Indian rivers (Subramanian 1987; Subramanian and others 1985a,b, 1987); sediments from Bay of Bengal (Sarin and others 1979), which receive the bulk of sediments delivered by Indian rivers; major Chinese rivers (Li and others 1984); world average river sediment (Martin and Meybeck 1979); and average soils (Bowen 1979).

The concentration of all the heavy metals considered in the Krishna River suspended particles are far in excess when compared to the world's average river suspended particles, average soils, and the two major rivers in China. The implication is that the pollution input (mainly due to industrial activities) of these metals in the Krishna River is significant as compared to the natural inputs. The average values in the Krishna River indicate that suspended particles are three to ten times richer than the bed sediments. Hence, the average computed for the Indian rivers, based on bed chemistry, does not represent the real situation, and it requires immediate revision based on the particulate chemistry for the entire subcontinent. The mean elemental concentration for all the measured elements in the Krishna River bed sediments appear to be of the same order of magnitude when compared to the Indian average.

Based on the annual sediment load in the Krishna River (Ramesh and Subramanian 1986), the annual heavy metal flux was calculated at various stations (Table 6). Similarly, based on the published values of mean annual sediment load and sediment chemistry, the average heavy metal flux for Godavari, Cauvery, Ganges, Brahmaputra, Indian, and world average sediments have been computed, and these data are also presented in Table 6 for comparison. In the Krishna River, the suspended load decreases sharply at the river mouth (Vijayawada). Decreases in the suspended load result from (1) building of dams and reservoirs, which trap sediments, (2) bank stabilization of rivers and (3) soil conservation practices (Berner and Berner 1987). Two major dams in between Srisaïlam and Vi-

Table 4. Correlation coefficients between elements in Krishna River basin^a

Element	Fe	Mn	Cr	Co	Ni	Cu	V	Zn	Pb
Fe	1								
Mn	0.93	1							
Cr	0.64	0.74	1						
Co	0.72	0.60	0.29	1					
Ni	0.67	0.68	0.84	0.29	1				
Cu	0.84	0.75	0.26	0.74	0.29	1			
V	0.73	0.68	0.36	0.49	0.30	0.78	1		
Zn	0.28	0.17	0.20	0.05	0.44	0.03	0.02	1	
Pb	0.25	0.17	0.16	0.20	0.47	0.05	-0.17	0.55	1

^aNumber of samples = 30 (16 for suspended and 14 for bed sediments).

Table 5. Comparative values ($\mu\text{g/g}$) of heavy metal composition of river-borne sediments

Element	Source of sediments ^a					
	Kr(SS) ¹ (n = 16)	Kr(BS) ¹ (n = 14)	Go(BS) ² (n = 26)	Ca(SS) ³ (n = 31)	Ga(SS) ⁴ (n = 4)	Br(SS) ⁴ (n = 3)
Fe	1,32,000	25,000	57,000	62,000	90,000	1,09,400
Mn	2,540	906	1,070	1,300	3,450	4,450
Cr	300	82	126	150	264	222
Co	300	32	47	100	223	168
Ni	180	32	51	150	137	179
Cu	220	35	82	60	252	108
V	630	140	300	300	—	—
Zn	270	26	54	500	1,800	916
Pb	45	—	11	40	—	—

Element	Source of sediments ^a					AS ⁹
	IA(BS) ⁵ (n = 128)	BB(SS) ⁶ (n = 12)	Ya(SS) ⁷ (n = 2)	Ye(SS) ⁷ (n = 1)	WA(SS) ⁸ (n = 10)	
Fe	29,000	39,000	55,500	32,000	48,000	40,000
Mn	605	529	1,055	800	1,050	1,000
Cr	87	84	83	72	100	70
Co	31	—	25	12	20	8
Ni	37	64	78	38	90	50
Cu	28	26	70	33	100	30
V	—	—	160	110	170	100
Zn	16	—	108	75	350	90
Pb	—	—	65	35	150	12

^aKr—Krishna; Go—Godavari; Ca—Cauvery; Ga—Ganges; Br—Brahmaputra; IA—Indian average; BB—Bay of Bengal; Ya—Yangtze; Ye—Yellow; WA—world average; AS—average soils.

SS—suspended sediments; BS—bed sediments; n—number of samples.

1—present study; 2—Subramanian (1987); 3—Subramanian and others (1985a); 4—Subramanian and others (1987); 5—Subramanian and others (1985b); 6—Sarin and others (1979); 7—Li and others (1984); 8—Martin and Meybeck (1979); 9—Bowen (1979).

jayawada have reduced the suspended load on the Krishna River by one-tenth (Ramesh and Subramanian 1986). In recent studies, Milliman and Meade (1983) point out that the suspended sediment loads transported to the oceans by the Nile and Colorado rivers have been reduced to nearly nothing by dams.

Similarly, dams and soil conservation have reduced the suspended load on the Mississippi and Zambezi rivers (Milliman and Meade 1983). The loss in suspended sediment transported to the oceans due to dams on large rivers, according to Milliman and Meade (1983) is about 0.5×10^9 t/yr or around 4% of the total river

Table 6. Heavy metal fluxes in suspended sediments of Krishna major rivers of Indian subcontinent and world average

Location	Sediment transport ^a ($\times 10^6$ t/yr)	$(\times 10^3$ t/yr)								
		Fe	Mn	Cr	Co	Ni	Cu	Zn	V	Pb
Krishna at										
Kolhar	0.5	0.11	2.31	0.18	0.18	0.12	0.24	0.14	0.53	0.03
Sangam	7.16	1.2	25.74	2.71	2.51	1.4	2.65	1.48	5.91	0.28
Raichur	14.86	1.87	37.73	3.88	3.98	2.01	3.73	2.87	10.46	0.24
Kurnool	16.86	2.19	42.93	4.27	6.39	2.65	4.43	1.8	11.68	0.47
Srisailam	11.26	1.26	19.38	2.47	2.57	1.16	1.96	1.5	10.48	0.17
Vijayawada	1.74	0.19	2.51	0.41	0.49	0.26	0.19	0.29	0.81	0.09
Tributaries										
Gataprabha	0.19	0.04	0.85	0.08	0.08	0.05	0.09	0.05	0.13	0.02
Malaprabha	0.75	0.08	1.39	0.16	0.16	0.14	0.09	0.18	0.29	0.04
Bhima	5.31	0.64	12.6	1.35	1.48	0.71	1.34	0.87	3.68	0.09
Tungabhadra	0.85	0.09	2.04	0.24	0.21	0.15	0.09	0.14	0.33	0.02
Mean	4.11 ^b	0.54	10.45	1.21	1.25	0.73	0.91	1.12	2.59	0.19
Godavari	170 ^c	9.71	181.9	21.42	7.99	8.67	13.94	9.18	50.49	1.87
Cauvery	0.71 ^d	0.04	0.92	0.11	0.07	0.11	0.04	0.36	0.21	0.03
Ganges	329 ^e	29.61	1,135.05	86.86	73.37	45.07	82.91	604.05	—	—
Brahmaputra	597 ^f	65.31	2,656.65	132.53	100.3	106.86	64.48	546.85	—	—
Indian average	1,212 ^d	35.15	733.26	105.44	37.57	44.84	33.94	19.39	—	—
World average	13,505 ^g	648.24	14,180.25	1,350.5	270.1	1,215.45	1,350.25	4,726.75	2,295.85	2,025.75

^aRamesh and Subramanian (1986).

^bRamesh and Subramanian (1988).

^cBiksham and Subramanian (1988).

^dSubramanian (1978).

^eAbbas and Subramanian (1984).

^fSubramanian (1979).

^gMilliman and Meade (1983).

suspended load. The metal flux of Krishna and Cauvery sediments appears to be low relative to other major rivers and global averages because of the low levels of sediment load. Within the basin, various fluxes indicate no uniformity, primarily because of different subbasin geology and various degrees of human impact.

Conclusion

There is considerable spatial variation in the heavy metal content in the Krishna River. Sediment particulates show heavy metal enrichment relative to the bed sediments. The proportion of different size populations in the sediment grains and human activities seems to account largely for heavy metal variations within the basin. Heavy metals distributed in various fractions (organic, carbonate, metal hydroxide, etc.) can be of value in assessing the potential impact of sediment resuspension upon water quality. Hence, further refinements can be made by studying the metal partitioning in the Krishna River sediments. All the metals considered show good correlation, indicating their cogenetic behavior within the basin. The average

bed sediment chemistry appears to agree well with the Indian average.

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