Processes Affecting the Transport of Materials from Continents to Oceans

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The major portion of the terrigenous material that reaches the estuarine and shallow marine environments is derived from the continents through weathering processes and transported via river systems. The products of continental weathering and erosion are transported in the form of suspended particulate matter, colloidal material, and dissolved species. The impact that these terrigenous materials have on the shallow water marine environment varies according to the nature and amount of the material and the site of discharge. Some materials introduced by rivers via an estuary may undergo major changes during their residence time in that environment before finally reaching the ocean. Others may be relatively unaffected during transit. The behavior of each material is dictated by its chemical composition and physical properties, and by the chemical, physical, and biological constraints imposed upon it by the estuarine environment. Understanding the chemistry of the estuarine and near-shore marine environment thus entails a knowledge of the chemical inputs into the environment and the nature of the interactions that take place among these materials within the environment. In river waters and in sea water the species Na^+ , Mg^{++} , Ca^{++} , K^+ , Cl^- , SO_4^{--} , HCO_3^- and H_4SiO_4 account for more than 98% of the total dissolved solids. These major elements determine the general chemical character of the waters. The other elements, the trace elements, are present in much smaller quantities, but may nevertheless play important roles in determining the chemical quality of the environment.

The sediments carried by rivers to estuaries and the near-shore marine environment are constituted primarily of the elements O, Si, Al, Ca, Mg, K, Na, and Fe with other elements present in trace amounts.

The continuum of reactions that occur between the waters and solids during the weathering process, transport through river systems and after sedimentation in the estuarine or coastal marine environment are a major influence on the composition of the aqueous phase and lead ultimately to the mineral assemblages

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that are preserved in the sedimentary record.

It may be instructive at this point to examine some of the types of reactions that occur during the sedimentary cycle. The materials of the earth's crust can be separated into two groups on the basis of chemical composition, the silicates and the carbonates. A third group, the evaporite-deposits, is quantitatively unimportant in terms of volume, but can play an important chemical role on a local scale. Chemical weathering of the earth's crust produces two types of products: a) dissolved species, and b) residual solids that are more stable under earth's surface conditions than the original minerals in the rocks being weathered. These residual solids are usually aluminosilicate minerals of the Phyllosilicate group (clays), or hydrous oxides of iron and manganese.

Present information suggests that, once formed in the weathering environment, little happens to the clay minerals other than shifts in the exchangeable ion population until they are deposited in the marine environment. Similarly, the alkali and alkaline earth cations freed in weathering reactions undergo few reactions other than minor involvement in ion exchange reactions with clay minerals and, perhaps with particulate organic matter, during their transit to the sea. In interstitial waters of estuarine and marine sediments, however, reactions occur that involve these elements. For instance, Drever (1) has reported exchange of Mg for Fe in clays in the sediments of the Rio Ameca basin. The clays take up Mg in exchange for iron, and the released iron reacts with sulfide ion in the anoxic interstitial waters to form iron sulfide. Depletion of Mg in the interstitial waters of deep sea sediments has also been reported (2, 3). The interstitial water environment in the sediments appears to be the locale in which significant chemical changes begin to occur.

The behavior of trace elements in estuaries is in marked contrast to the nearly conservative behavior of the major ions. Trace elements participate in a variety of biogeochemical reactions, and an understanding of these reactions is crucial to the understanding of the cycling of trace elements between continents and oceans.

River Transport

River input is the major source of trace elements to the estuarine and near-shore marine environments. The chemical form of an element in river water to a great extent determines the type of reaction it participates in during its passage through an estuary. Trace elements can be transported by rivers in several different fractions:

A. Dissolved

1. Inorganic - includes the "free" hydrated ion and inorganic ion pairs (i.e., FeOH^{2+} , FeCl_2^+)

2. Organic - composed of trace elements complexed with dissolved organic matter. Trace elements in the dissolved fraction are highly reactive and readily participate in all types of estuarine processes.

B. Particulate

1. Adsorbed on the surface of particles - most suspended particles in natural waters are negatively charged and cations are attracted to their surfaces. Additionally trace elements are adsorbed on ion exchange positions of clay minerals and on hydrous oxides of iron and manganese. Both kinds of adsorbed ions are easily desorbed from particles when the chlorinity of surrounding solution changes.

2. Coprecipitated with iron and manganese in hydrous oxide coatings - during weathering, iron is released to solution and immediately precipitates (at pH's greater than 4-4.5) as a hydrated oxide. This material readily scavenges trace metals either by coprecipitation or sorption. Manganese in solution commonly coprecipitates with iron. These fine-grained, high surface area hydrous oxides coat detrital particles and are transported with them by rivers (4). The coatings are stable in highly oxic surface waters but are unstable under anoxic conditions and quickly dissolve releasing the contained trace metals.

3. Precipitated on the surface of detrital particles - some trace metals can precipitate in the form of pure phases on surfaces of clay minerals or other detrital particles at low temperatures and pressures (5). These precipitates can dissolve under varying conditions of pH and pE.

4. Bound within the crystalline lattice of sediment particles - these trace metals can only be released to solution under harsh chemical conditions rarely encountered in natural waters.

5. Bound within organic particulates - stable in oxic environments and unstable in anoxic waters and sediments. Rate of decomposition and subsequent release of trace metals depends on the composition of the organic matter and the intensity of bacterial activity.

The chemical reactivity of trace elements is different in each of these fractions; and consequently, the behavior of an element passing through an estuary is strongly influenced by its distribution among these fractions. Gibbs (6) investigated the partitioning of trace metals among some of these fractions in the Amazon River System. His results illustrate the substantial variation in the distribution of four elements among these fractions during their transport in the river (Figure 1). In the Amazon, most of the Fe and Mn are transported as oxide coatings, while most of the Cu and Co reside in the crystalline lattice. In addition, large portions of the total Cu and Mn are in the dissolved state and significant amounts of Fe and Co are bound with organic particulates. Gibbs also reported that the order

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of dominance of the fractions (i.e., Fe: coating > organic > solution) varied little between rivers but that the absolute percentage of an element in each fraction did change. Thus, since the chemical reactivity of elements varies between the fractions, the amount of each trace metal available for different biological and geochemical reactions in estuaries varies from river to river and must be determined for each element under study.

Several workers have found that trace element concentrations vary greatly between different rivers (7, 8, 9, 10). This observation is not unexpected since the concentration in a river depends on the relative concentration of trace elements being weathered in the drainage basin and on the rate of weathering of the different minerals containing trace elements. Recent studies have also shown that trace metal concentrations in rivers vary greatly as a function of discharge rate, suspended load and time of year.

Carpenter and Grant (11) completed an extensive study of the temporal variability of trace metals transported by the Susquehanna River to the Chesapeake Bay. They sampled the river on a weekly basis for a period of approximately $1\frac{1}{2}$ years at a station one mile below the Conowingo Dam. Each sample was separated into three fractions and these fractions were analyzed for trace metals:

1. Settled solids (SS) - 100 liters of river water were allowed to settle for 10-14 days. The supernatant water was removed and the remaining slurry was digested with 1 M HCl and 1.5 M HAc for 48 hours at 60°.

2. Filtered solids (FS) - 50 liters of the supernatant water was filtered through acid-washed 0.2 μ m cellulose acetate membrane filters. The filtered solids were then treated identically to the settled solids. The remaining 50 liters of supernatant water was centrifuged to determine the weight concentration of solids.

3. "Soluble" (SOL) - 50 liters of the filtrate from step two was run through a cation exchange resin (Chelex 100 in acid form). The resin was eluted with 1 M HCl and the eluate was evaporated to dryness. The residue was oxidized with nitric acid and made to volume with 0.1 M HCl.

All three fractions were then analyzed for their trace metal content by atomic absorption spectrophotometry preceded by APDC complexation and MIBK extraction. The standard deviation of replicate samples varied between 1.0 and 2.2%.

The discharge of water and suspended solids by the Susquehanna River over a 17 month period in 1965-66 is depicted in Figure 2. The most striking feature of the data is the large variation of both discharges during the year. As we would expect both discharges are greatest during early spring. In fact 5.1% of the total annual discharge of solids occurred on one day, February 17; and 30% of the total annual solids discharge

occurred in one week, February 14-20. Because of these large fluctuations in discharge rates, we conclude that estimates of total solids and total trace element discharge of rivers computed from concentrations and flow rates determined during periods of low or moderate discharge are not representative of the true total annual discharge.

As a first approximation the trace metal concentration ($\mu\text{g/kg}$ water) data correlates well with the solids discharge (Figures 3, 4 and 5). The concentrations are highest in the spring and lowest in the summer and fall. Closer inspection, however, reveals differences in the patterns of particular elements and total solids. Mn, Ni, Zn and Co exhibit large concentrations in January, and Cu, Cr and Mn have concentration maxima in late spring or early summer. The temporal variability of the solids discharge can be eliminated from the trace metal data by computing the weight concentrations of the metals in the solids fraction (mg/kg solids). This data is shown in Figures 6 and 7. Generally there are peaks in concentration for all the metals during December and January and secondary peaks for Co, Cr, Ni, Cu and Mn in July. Since decaying organic matter (particulate and dissolved) is abundant in the Susquehanna during these two periods, the high concentrations may be the result of binding of the metals to particulate organisms. Figures 8 and 9 illustrate the distributions of Fe, Mn, Cu, Ni and Zn among the three fractions during the 17 month sampling period. Most of the Fe and Zn are associated with the solids throughout the year except during January when a substantial portion of the two metals are in the dissolved state. The partitioning of Mn between solution and solids varies throughout the year. There is a substantial amount of soluble manganese present during January and early spring, but the rest of the year manganese is associated predominantly with the solids fractions. Large amounts of Ni and Cu are in the dissolved form throughout the year but the ratio of the soluble to that associated with the solids fraction increases in December and January. We believe that the high concentrations of dissolved trace metals during early winter can be associated with the increased amounts of decaying organic matter in the river during this period. Further investigation is needed to confirm this conclusion.

The importance of frequent sampling in the estimation of trace element transport is pointed out in Table 1. Turekian's estimates are based on one sampling period during the summer of 1966, and those of Carpenter and Grant are time averages over a 17 month period. The discrepancies between the two sets of data are greater than an order of magnitude for many of the metals. The higher estimates of Turekian may be attributed to the high concentrations of several trace metals in the solids fractions noted by Carpenter during July, 1966 (Figures 6 and 7).

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Table 1. Trace metal transport of the Susquehanna River (Tons)

	Carpenter et al. (11)	Turekian & Scott (7)
Suspended solids	767,000	300,000
Fe	43,000	---
Mn	5,300	120,000
Zn	650	---
Ni	215	3,000
Cu	105	---
Co	88	1,500
Cr	53	870
Cd	2	---
Ag	---	45
Mo	---	97

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Although rivers are the dominant source of trace metals entering estuaries, rainfall and particulate fallout may be important sources in specific locales. Particularly for volatile trace metals, such as Se, Hg and Pb. For example Patterson (12) has reported that dry aerosol deposition of Pb in the Southern California Bight equals more than 75% of the amount of Pb transported by rivers and waste waters. Similarly, in the Delaware estuary, Biggs (13) has found that the concentration of Cd and Pb in rainfall is higher than the concentrations observed in unpolluted streams discharging into the estuary.

Biogeochemical Processes in Estuaries

Once the rivers have transported trace elements to an estuary, the elements can undergo several kinds of physical, chemical and biological reactions. It is important to identify these reactions since estuarine processes affect the amount and the form of the trace elements transported to the ocean. On the basis of their behavior in estuaries, elements can be classified as either conservative or non-conservative. Conservative elements display a linear relationship between concentration and chlorinity along the length of an estuary which reflects a simple, physical mixing process between river and sea water. Conservative behavior is typically exhibited by the major ions and some minor constituents, such as silica in the Merrimac estuary (14, and Figure 10). Most trace elements are nonconservative, like iron (Figure 10), and physical mixing processes alone cannot account for their distributions in an estuary.

There are five major categories of reactions which influence trace metal transport in estuaries:

1. Flocculation and sedimentation - these processes are encouraged by the increased salinity of estuarine waters. As a result of the salt wedge type of circulation commonly observed (Figure 11) sedimentation is concentrated in the upper reaches of estuaries. Suspended sediment discharged by rivers is carried

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50 toward the ocean by the fresh water at the surface of the estuary.
49 As the solid particles settle, they fall into the salt wedge
48 layer moving toward the mouth of the river. The particles con-
47 tinue to settle and sedimentation is concentrated in the low
46 velocity region near the tip of the salt wedge. Schubel (pers.
45 comm.) estimates that, in the Chesapeake Bay 90% of the solids
44 discharge is deposited in the northern third of the estuarine
43 system. His argument is substantiated by the distribution of Fe
42 in sediments along the length of the bay. The time-averaged
41 concentration of Fe in suspended solids discharged by the Susque-
40 hanna River is about 1000 PPb (11). The concentration of Fe in
39 northern Chesapeake Bay sediments averages 5%, while the concen-
38 tration in southern bay sediments averages 3%. These data sug-
37 gest that most of the sediments of the southern bay originate
36 from shoreline erosion rather than from river discharge.

35 2. Mineral-water interaction - the rapid changes in salin-
34 ity, pH and oxygen content of estuarine waters can affect the
33 precipitation or dissolution of minerals (for instance, the
32 dissolution of the hydrous oxide coatings under low O₂ conditions
31 in water or sediments).

30 3. Adsorption/desorption - due to the high concentration of
29 Na and Mg in seawater, ion exchange reactions occur rapidly in
28 estuarine waters as sediment particles adsorb the seawater cati-
27 ons and desorb trace elements.

26 4. Diagenesis and remobilization of trace metals in
25 sediments - the decreased pH and strong pE gradient in sediment
24 interstitial waters brought about by the decomposition of organic
23 matter significantly affects the solubility of trace element
22 solid phases and ion exchange equilibria. Trace metal concen-
21 trations in sediment pore waters are typically orders of magni-
20 tude greater than in the overlying water, and the flux of trace
19 elements across the sediment-water interface may constitute an
18 important mass transfer process in estuaries.

17 5. Biological processes - interactions with aquatic
16 organisms may strongly influence the behavior of trace elements
15 during their passage through estuaries. The generally recognized
14 mechanisms are: ingestion of particulate suspended matter con-
13 taining trace metals, trace metal complexation with organic
12 chelates produced by organisms, trace element uptake in metabolic
11 processes, and ion exchange and sorption on the surfaces of
10 organisms (15).

9 Most trace elements undoubtedly participate in all these
8 major categories of reactions to some extent, and it is important
7 to identify which reactions most strongly influence the transport
6 of each element under study. To this end, our group at Johns
5 Hopkins has concentrated on the effect of sediment-water exchange
4 processes on trace element cycling.

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Desorption of Zn from Estuarine Sediments

Bradford (16) investigated the influence of sediments on zinc concentrations in the overlying water in Chesapeake Bay. Using anodic stripping voltammetry, he observed large Zn concentrations just above the sediment-water interface in the northern bay during May. The distribution of total zinc (UV oxidized) and inorganic zinc (raw sample) just above the sediment-water interface at station 914S is shown in Figure 12. During May there is a large concentration of zinc at this station. The zinc is totally in the inorganic form because there is no difference in concentration between the oxidized and raw samples. The only other major change occurring during May at 914S was a rapid increase in the chlorinity from less than 0.2 ‰ to greater than 3 ‰ (Figure 13). The large zinc concentration cannot be the result of mixing of bay water and river water since the concentration of zinc never exceeded 6 ppb in either of these waters at this station. Thus, the large increase in Zn must have come from sediments. This conclusion is substantiated by the concentration gradients of Zn at station 914S during April, May and June (Figure 14). During April Zn varies from 4.5 ppb at 4 m to 5.5 ppb at 1 m above the interface. In May there is a large vertical concentration gradient which implies a flux of Zn from the interface. In June the Zn concentration is uniform at this station. We conclude from these data that zinc was desorbed from freshly deposited river sediments at this station as Mg and Ca from the salt wedge were adsorbed. During March and April the Susquehanna discharges a large quantity of suspended solids to the upper reaches of the Chesapeake Bay. These solids form a layer 1-2 cm thick in the northern bay. The ion exchange positions of these solids are in equilibrium with the river water ratios of the major cations. Once the river discharge subsides the salt wedge begins to move landward again. During May the salt wedge moved across the sediments at 914S, and in response to the ion exchange equilibria, the sediments adsorbed the major sea water cations and desorbed Zn to the overlying water. This conclusion confirms earlier observations suggesting that Zn rapidly desorbs from surface sediments as the chlorinity increases above 0.4‰ (17, 18).

Remobilization of Trace Metals in Sediment Pore Waters

In the highly oxygenated conditions of normal river beds the hydrous Fe and Mn oxide layers coating sediment particles are stable. When the detrital particles reach the marine environment and become incorporated into the organic rich bottom sediments of an estuary or marine delta, they encounter very different conditions. Oxygen is usually depleted within the upper few centimeters of sediment by bacterially mediated oxidation of organic material. In brackish and marine waters, after oxygen is

depleted, bacteria use sulfate as an electron acceptor to continue the oxidation of organic matter (equation 1):

These reactions decrease the sulfate concentration and pH and increase the concentrations of bicarbonate, phosphate and sulfide. Additionally strongly reducing conditions are produced in which the oxide coatings on detrital particles are chemically unstable. Solution of the oxide coatings produces large concentrations of dissolved iron, manganese and other trace metals in the interstitial waters. The vertical distribution of dissolved Fe(II) and Mn in the interstitial waters of the upper meter of sediments at a station in the northern Chesapeake Bay is shown in Figure 15. The concentrations of both Fe and Mn increase rapidly 1 to 2 cm below the sediment water interface as a result of the dissolution of the oxide coatings. Below 10 cm, the concentrations decrease rapidly and become nearly constant below 40 cm. We attribute the rapid decreases of the concentrations to the precipitation of authigenic trace metal phases in the interstitial water as phosphate and carbonate increase rapidly with depth. Siderite and vivianite control the dissolved iron concentrations below 10 cm (19) while rhodocrosite controls the Mn distribution (20).

The concentrations of other trace metals which had coprecipitated with Fe and Mn during weathering also increase as the oxide coatings dissolve. In northern Chesapeake Bay pore waters, Cu and Pb mimic the distributions of Fe and Mn (Figure 16). Thus just below the sediment-water interface there is a concentrated reservoir of dissolved trace metals which can be transferred across the interface by molecular diffusion, sediment resuspension and bioturbation (21). Because of the magnitude of the trace metal concentrations in sediment pore waters, very little transfer would have to occur to significantly alter the overlying water concentrations, and consequently, to increase trace metal transport to the oceans.

Conclusions

At the present time we can draw a fairly complete picture of the transport of major ions between continents and oceans, due to their nearly conservative chemical behavior. On the other hand, trace elements are non-conservative species in estuaries, and extensive spatial and temporal surveys of their distributions are required to delineate the modes of transport of individual elements. Specifically, we need the following kinds of data:

1. River transport data for all important trace metal fractions to determine season-to-season and year-to-year variations.
2. Data on the flux of trace elements through biological systems

3. Quantitative assessment of the role of sediments in trace metal transport.

4. Data on oceanic input of trace elements into estuaries so that estimates of net transport to the ocean can be calculated.

In general we know the kinds of reactions which are important in trace metal transport. However, in many cases we do not know which reaction is most important for a given element, and in almost all cases we do not know the amount of each element participating in the reaction or the rate at which the reaction is occurring. The data that has been collected and that we are now collecting will help to remedy this situation.

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Fig. 1. Mechanisms of metal transport in the Amazon River.

Fig. 2. Susquehanna River discharge, water and suspended solids, April 1965 to September 1966 (after Carpenter et al (11)).

Fig. 3. Concentrations of Cd, Cr, and Co (mg/kg H₂O) in Susquehanna River discharge, April 1965 to September 1966 (after Carpenter et al. (11)).

Fig. 4. Concentrations of Cu, Ni and Zn (mg/kg H₂O) in Susquehanna River discharge, April 1965 to September 1966 (after Carpenter et al. (11)).

Fig. 5. Concentrations of Fe and Mn (mg/kg H₂O) in Susquehanna River discharge, April 1965 to September 1966 (after Carpenter et al. (11)).

Fig. 6. Concentrations of Ni, Zn, Mn and Fe (mg/kg solids) in Susquehanna River discharge July 1965 through July 1966 (after Carpenter et al. (11)).

Fig. 7. Concentrations of Cd, Cr, Co and Cu (mg/kg solids) in Susquehanna River discharge July 1965 through July 1966 (after Carpenter et al (11)).

Fig. 8. Partitioning of Mn and Fe between solution and solids in Susquehanna River discharge, April 1965 to September 1966 (after Carpenter et al (11)).

Fig. 9. Partitioning of Cu, Ni and Zn between solution and solids in Susquehanna River Discharge, April 1965 to September 1966 (after Carpenter et al (11)).

Fig. 10. Conservative behavior of silica vs. non-conservative behavior of Fe in the Merrimac River estuary (after Boyle et al (14)).

Fig. 11. Diagrammatic sketch of an estuarine salt wedge (after Neihsel (22)).

Fig. 12. Near bottom Zn concentrations with time at Station 914S in the upper Chesapeake Bay (after Bradford (16)).

Station 914S

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The chlorinity with depth and time at Station 914S.

Fig. 13. The chlorinity with depth and time at Station 914S in the upper Chesapeake Bay (after Bradford (16)).

Fig. 14. Near bottom Zn concentrations in April, May and June at station 914S in the upper Chesapeake Bay (after Bradford (16)).

Fig. 15. Concentrations of dissolved Fe and Mn as a function of depth beneath the sediment-water interface.

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Fig. 16. Concentrations of dissolved Pb and Cu as a function of depth beneath the sediment-water interface.