

Transport of major solutes and the relationship between solute concentrations and discharge in the Apure River, Venezuela

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Abstract. The Apure River is a major white-water tributary of the Orinoco River in Venezuela. The Apure is rich in solutes; its contribution to dissolved inorganic solids in the Orinoco (24%) is proportionately much greater than its contribution to discharge (6%). About 40% of the calcium and bicarbonate at the mouth of the Orinoco originate in the Apure drainage. The relationship between discharge and the concentrations of major solutes in the Apure was characterized with a two-compartment hyperbolic mixing model. Previous applications of the two-compartment model have been based on separate determinations of the model parameter β , which is a constant describing watershed hydrology, for each solute from data on concentrations. The use of a weighted mean β for all solutes is proposed as a means of assessing the importance of processes other than mixing. The model, when used on the Apure data, shows that a strong dilution effect prevails for sodium, calcium, magnesium, sulfate, and bicarbonate, and that a strong purging effect (increase of concentration with increasing discharge) is characteristic of soluble silicon. Biological immobilization of soluble silicon by diatoms during the season of low discharge is sufficiently large to account for the positive relationship between discharge and the concentration of soluble silicon. Specific transport rates of solutes from the basin are generally higher than global averages. In contrast, specific transport of chloride is low. Atmospheric sources control chloride transport in the Apure watershed; the low transport rates of chloride are probably explained by the great distance between the Apure watershed and the oceanic sources of atmospheric chloride.

Introduction

The Apure River in western Venezuela is a major white-water tributary of the Orinoco River. The headwaters of the Apure are in the Venezuelan Andes, but most of the watershed is a vast alluvial plain called the Llanos, which supports a tropical savanna. River discharge is highly seasonal in response to alternating wet (April–November) and dry (December–April) seasons. Seasonal trends for particulate matter and for carbon, nitrogen, and phosphorus are closely related to the hydrologic cycle (Saunders &

Lewis 1988a). Concentrations of dissolved nutrients tend to be positively correlated with discharge. Particulate matter reaches a peak of concentration with the first sharp rise in water level at the beginning of the wet season. The Apure is rich in dissolved solids (Lewis & Saunders 1989) and contributes a disproportionately high load of sediment to the Orinoco River (Meade et al. 1989).

In this study we summarize the major ion chemistry of the Apure River over a two-year period and assess its influence on the chemistry of the lower Orinoco. Seasonal variations in concentration are examined in relation to discharge by means of a two-compartment hyperbolic mixing model (Johnson et al. 1969). These data characterize major solute yields and provide insight into biogeochemical processes of tropical savanna, a major ecosystem type about which relatively little is known. Detailed site descriptions have appeared elsewhere (Saunders & Lewis 1988a, b).

Methods

Water samples were collected at four-week intervals from March 1984 through November 1985 near the mouth of the Apure River. The samples were collected with a flow-proportional, depth-integrating sampler (Nordin et al. 1983) at three points across the channel. Water from the three points was combined to make a single composite sample. Samples were kept cool and dark and were filtered with glass-fiber paper ($0.7\ \mu\text{m}$ effective pore size: Sheldon 1972), usually within 12 hours of collection. A subsample of filtered water was acidified ($\text{pH} < 2$) for major cation analysis by atomic absorption spectrophotometry. Lanthanum was added as a releaser and cesium was added to suppress flame ionization (Rains 1982). A separate, untreated subsample of filtered water was retained for analysis of Cl and SO_4 by ion chromatography. Bicarbonate was measured by potentiometric incremental titration of unfiltered water by the method of Gran (Stumm & Morgan 1981). Soluble silicon was determined by a colorimetric test based on molybdate (Armstrong & Butler 1962).

Stage height measurements were available on a daily basis at San Fernando de Apure. The relationship between stage height and discharge has been presented previously (Saunders & Lewis 1988a). Existing calibration measurements of discharge (MOP 1972) show no hysteresis; discharge in the Apure thus conforms to an assumption critical for use of standard mixing models (Hall 1970).

Transport was calculated for each day of both years as the product of discharge and concentration. Discharge data were available for each day,

and concentration data for days not sampled were estimated by linear interpolation. For months not sampled (January–February 1984 and December 1985), values from the other year were substituted. These months fell chiefly during the season of low water when concentrations were relatively stable. Annual transport was calculated as the sum of daily transport for each year. Discharge-weighted mean concentration for each solute was estimated as annual transport divided by annual discharge. An area of 122,000 km² for the Apure watershed above San Fernando de Apure was used in computing specific transport.

Results

Patterns of concentration

Discharge-weighted mean concentrations for the major solutes in the Apure River are presented in Table 1. The dominant cation was calcium (63% as μeq) and the dominant anion was bicarbonate (79% as μeq). The mass ratios of the major ions ($\text{Na}/(\text{Na} + \text{Ca}) = 0.20$; $\text{Cl}/(\text{Cl} + \text{HCO}_3) = 0.03$), and the sum of inorganic solutes (111 mg/L) place the Apure in the rock dominance category described by Gibbs (1970). The ranking of ions ($\text{Ca} > \text{Mg} > \text{Na} > \text{K}$; $\text{HCO}_3 > \text{SO}_4 > \text{Cl}$) on the basis of $\mu\text{eq/L}$ conforms to the pattern characteristic of nearly half of the rock-dominated surface waters in the world (Meybeck 1980). The charge balance for the major ions was nearly exact (Table 1); a slight excess of anions (0.9%; $[\text{cations} - \text{anions}]/[\text{cations} + \text{anions}]$) occurs when dissolved organic carbon is considered (31 $\mu\text{eq/L}$ given a discharge-weighted mean DOC con-

Table 1. Concentrations of major ions in the Apure River over a two-year period.

	Discharge-weighted mean concentration		Range, mg/L
	mg/L	$\mu\text{eq/L}$	
Na	4.1	178	2.8–8.0
K	2.2	55	1.5–2.6
Ca	16.2	811	11.9–30.4
Mg	3.0	246	2.4–5.6
Cl	1.7	47	0.6–5.4
SO ₄	10.7	223	5.0–34.7
HCO ₃	61.8	1013	49.0–110.0
Si	5.3	—	2.2–6.3

centration of 6.2 mg/L and assuming 5 μ eq/mg DOC: Saunders & Lewis 1988a; Lewis & Saunders 1988). For individual dates, when DOC is included, the maximum imbalance showed a 13.5% excess of anions; 80% of dates balanced within 5%, and 60% balanced within 1%.

Temporal trends for the major solutes were compared by correlation analysis (Table 2). The pattern for K, which exhibited little variation over time, was unrelated to that of any other major solute and was largely independent of discharge. All correlations between Si and other solutes except K were negative and most were significant at $p = 0.01$. Correlations among pairings of all other major solutes were significant and positive. Correlations between discharge and concentrations of solutes except K were significant.

The high frequency of significant correlations among concentrations of the major solutes is probably caused by the strong influence of discharge on concentration. The linear effects of discharge can be removed statistically by partial correlation for each pair of solutes with discharge as a controlling variable. Some important relationships independent of discharge are evident from the partial correlation coefficients (Table 2). A strong relationship existed among Ca, Mg, and HCO_3 , and between Cl and SO_4 . Correlations among the divalent cations and HCO_3 probably reflect common geologic sources. A strong relationship between Cl and SO_4 might be expected where atmospheric input dominates the sources of these ions in a watershed, as it does for the nearby Caura River (Lewis et al. 1987). Although Cl inputs were probably atmospheric, as explained below, sulfate concentrations were far too high to be explained exclusively by atmospheric sources.

For most of the ions, concentration was inversely related to discharge. A strong dilution effect prevailed for Na, Ca, Mg, SO_4 , and HCO_3 (Fig. 1);

Table 2. Simple correlations (Pearson product-moment) among the major ions and discharge (above diagonal) and partial correlations among major ions after controlling for discharge (below diagonal). All variables were log-transformed except discharge, which was expressed in the form $1/(1 + \beta Q)$.

	Cl	SO_4	HCO_3	Na	K	Ca	Mg	Si	$1/(1 + \beta Q)$
Cl		0.81*	0.71*	0.74*	-0.27	0.72*	0.63	-0.51	0.61*
SO_4	0.72*		0.82*	0.84*	-0.34	0.87*	0.80*	-0.66*	0.88*
HCO_3	0.49	0.12		0.89*	-0.55	0.96*	0.94*	-0.63*	0.91*
Na	0.58*	0.21	0.40		-0.38	0.90*	0.94*	-0.59*	0.91*
K	0.04	0.22	-0.20	0.16		-0.31	-0.30	0.43	-0.49
Ca	0.50	0.37	0.76*	0.49	0.27		0.95*	-0.53	0.88*
Mg	0.24	-0.04	0.66*	0.70*	0.35	0.78*		-0.54	0.90*
Si	-0.13	-0.07	0.06	0.23	0.14	0.31	0.36		-0.72*

* 2-tailed $p < 0.01$.

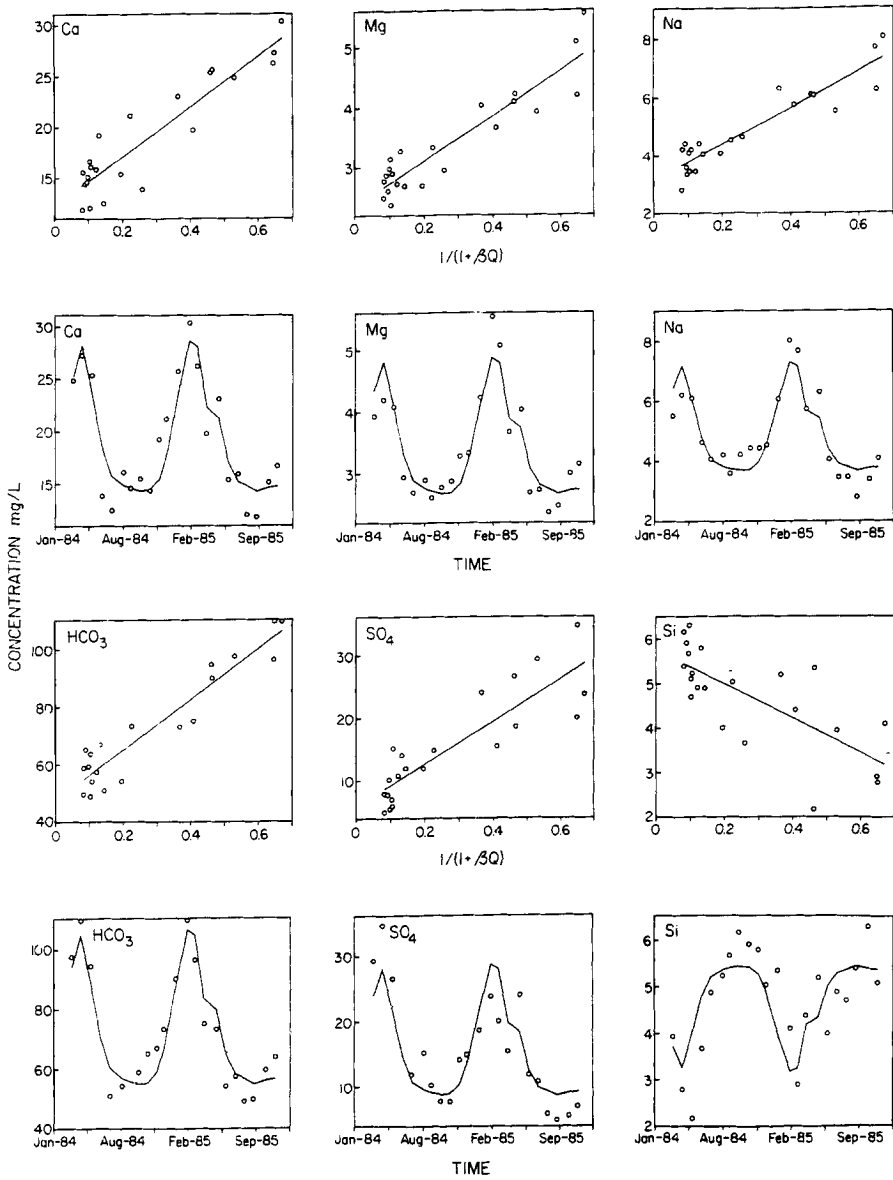


Fig. 1. Relationship between concentration and discharge for eight major solutes in the Apure River. The top row shows concentrations against a hyperbolic function of discharge ($1/[1 + \beta Q]$) for cations; the second row shows concentrations over time for cations. Row three and four give similar information for anions and silicon. Data points are based on sampling; solid lines are concentrations predicted by the two-compartment model using the weighted mean β . Potassium and chloride were omitted due to their poor fit to the model.

chloride showed a less pronounced dilution effect. Seasonal variations in potassium concentration were relatively small and bore little relationship to variations in discharge. Silicon was unique among the major inorganic solutes in the Apure River because it showed purging (a positive relationship between concentration and discharge). Because silicon is derived from weathering, a purging effect was unexpected.

The relationship between concentration and discharge in the Apure River was characterized for the measured values of each solute by a two-compartment hyperbolic mixing model (Johnson et al. 1969). The mixed concentration, C , of a solute in the river is defined as:

$$C = (C_0 V_0 + C_\alpha V_\alpha) / (V_0 + V_\alpha).$$

V_0 is the initial volume of water in the system and V_α is the volume of water that is added through storm events. C_0 and C_α are the corresponding concentrations. River discharge (Q) is proportional to the added volume:

$$V_\alpha = rQ$$

where r is the residence time of the added volume. By substituting rQ for V_α , and a constant β for r/V_0 , C can be defined as:

$$C = C_\alpha + (C_0 - C_\alpha) / (1 + \beta Q).$$

River concentrations (C) will vary in response to changes in V_α ; at low discharge, $C = C_0$; at maximum discharge, C approaches C_α .

The two-compartment hyperbolic model uses three parameters to describe solute concentration as a function of discharge:

- a hydrologic characteristic of the watershed (β),
- solute concentration in the added volume (C_α), and
- solute concentration in the initial volume (C_0).

It is common practice to select an initial value for β and then to solve for the other two parameters by linear regression; the fit is improved iteratively by testing different values for β (Johnson et al. 1969; Lesack et al. 1984). Nonlinear regression has also been used to solve for all three parameters simultaneously (Hall 1970; Saunders & Lewis 1988a). All three parameters are estimated separately for each solute.

Table 3 shows the fit of the Apure River concentration data to the two-compartment hyperbolic mixing model as obtained by nonlinear regression. A good fit was obtained for all solutes except K and Cl. However, the model predictions of C_0 for solutes characterized by a strong dilution

Table 3. Model parameters for the two-component hyperbolic model.

Variable	Variable β model			Fixed β model ⁽¹⁾		
	$\beta \times 10^4$	95% confidence interval	r^2	r^2	C_x	C_0
Na	25.3	-7.7 to 58.3	0.86	0.86	3.2	9.3
K	11.6	-53.8 to 76.9	0.23	0.22	2.3	1.6
Ca	24.7	-9.8 to 59.2	0.84	0.84	12.3	36.7
Mg	55.4	-34.6 to 145.5	0.86	0.85	2.4	6.1
Cl	6.6	-28.3 to 41.6	0.33	0.31	1.3	4.1
SO ₄	6.7	-6.3 to 19.7	0.78	0.76	5.9	39.9
HCO ₃	38.3	-11.2 to 87.8	0.90	0.89	47.5	135.5
Si	2.9	-8.1 to 14.0	0.55	0.53	5.8	1.9

⁽¹⁾ $\beta = 0.002547$, weighted mean

effect (Ca, Mg, Na, HCO₃, SO₄) exceeded the average concentrations measured over intervals when river flow consisted entirely of the base flow component (i.e., when C should equal C_0). The consistently large positive deviation of predicted from observed C_0 for these ions indicates a deficiency in the model or a failure to meet all modelling assumptions.

Despite the good fit of the data for most solutes to the two-compartment hyperbolic model, estimates of the parameter β varied greatly among different solutes. Because β represents a specific hydrologic characteristic of the watershed, it should be constant; estimates should not vary among the solutes unless concentrations are affected by processes that overshadow simple mixing. High variability of β as estimated from concentration data has been observed in other studies (Lesack et al. 1984; Saunders & Lewis 1988a). This variability suggests either that the model is conceptually unsound, or that the simultaneous estimation of β , C_0 , and C_x from concentration data causes high uncertainty in β . By nonlinear methods, Saunders & Lewis (1988a) have shown that statistical uncertainty in β can be very high for the fit of three parameters to concentration data, even though the amount of variance accounted for is high. Table 3 shows the broad confidence limits for β as determined from the Apure data. It is clear that a major contribution to variation in β from one solute to another is simply the statistical uncertainty in β for any given solute.

Statistical uncertainty in the parameter estimation of the two-compartment model can best be remedied by a more secure determination of β , followed by a two-parameter estimation of C_0 and C_x from the concentration data. An improved estimate of β can be obtained by averaging values from the eight separate solute models. Because the fit varies widely among the

solutes, it would not be appropriate to give equal weight to all solutes. β values for all solutes are included here, but each is weighted by its r^2 value. The contribution of solutes that fit the model poorly is thus negligible. The weighted average for β , $0.002547 \text{ s} \cdot \text{m}^{-3}$, falls within the 95% confidence for all individual estimates except SO_4 and Si (Table 3).

The weighted mean β was applied as a constant to each relationship between solute concentration and discharge. A single value of β for all solutes is implicit in the model and establishes a uniform basis for comparison of solutes. The resulting models have only two parameters and these can be estimated by linear regression techniques. A reduced closeness of fit of the data to the model is expected when two parameters are allowed to vary instead of three (Table 3). The fit for individual solutes is not very sensitive to the selection of β , however; an order of magnitude change in β will have little effect on fit.

Patterns of transport

Annual transport of major solutes from the Apure watershed is shown in Table 4, which also provides a comparison with the total transport in the Orinoco River near its delta. Even though the Apure accounted for only 6% of annual flow in the Orinoco, it contributed 24% of inorganic solutes. About 40% of the annual transport of Ca and HCO_3 at the mouth of the Orinoco originated in the Apure drainage.

The relationship between transport (T) and discharge (Q), when modeled as a power function ($T = aQ^b$), showed the expected dilution effect ($b < 1$)

Table 4. Transport of major ions from the Apure River watershed and the relationship between transport and discharge, 1984–85. Seasonal contributions of the Apure to the Orinoco are based on two years of data.

	Transport, tons/day, Apure	Contribution to Orinoco, %			Transport-discharge relationship		
		Annual	Low-water	High-water	b	S.E. b	r^2
Na	677	16.3	8.3	18.2	0.770	0.023	0.98
K	357	17.0	6.5	19.5	1.066	0.027	0.99
Ca	2,683	38.0	18.5	42.3	0.769	0.027	0.98
Mg	493	28.2	13.1	32.2	0.806	0.020	0.99
Cl	273	13.3	5.9	17.3	0.713	0.088	0.77
SO_4	1,769	27.3	19.6	30.1	0.562	0.057	0.84
HCO_3	10,204	40.3	18.0	46.1	0.783	0.022	0.90
Si	857	10.0	2.8	12.5	1.178	0.038	0.98
Discharge	–	5.8	2.5	6.5	–	–	–

for all major solutes except K and Si (Table 4). Potassium concentration varied little over time and thus the exponent is close to 1. Because silicon concentration showed a purging phenomenon, the exponent is greater than 1.

Solutes from the Apure comprised a greater portion of transport in the Orinoco during high water than during low water (Table 4). For most solutes, seasonal variations in contributions of the Apure to solute transport in the Orinoco were parallel to those of discharge, in part because the seasonal patterns of concentration were similar in the two rivers (cf. Lewis & Saunders 1988). For SO_4 and Si, however, the seasonal differences were more exaggerated than would have been expected from discharge, and in both cases the amplitude of variation in the Orinoco was much smaller than that in the Apure.

Discussion

The parameter β from the widely used mixing model of Johnson et al. (1969) describes hydrologic features of the watershed that should be constant and independent of solute concentrations. Heretofore, β has been derived separately for each solute, which produces as many estimates of β for a given watershed as there are solutes. Biogeochemical processes have been used to explain variation in β among solutes, but such interpretation may often be unwarranted in view of the uncertainty associated with estimates of β from the three-parameter model, and in the absence of any standard to which individual estimates of β could be compared.

We have suggested a simple mechanism to obtain a single value for β and thus establish a common basis for assessing pattern in the comparison of observed and expected concentrations for each solute. The use of fixed β makes it easier to identify solutes for which biogeochemical processes cause deviations from the pattern expected when two-compartment mixing is the dominant process. Apure data for K and Cl, for example, did not conform well to the mixing model. This result would indicate important biogeochemical influences on Cl and K, in contrast to other ionic solutes. Hamilton & Lewis (1987) found that these two solutes did not behave conservatively in a lake on the Orinoco floodplain. Biological dynamics may overshadow mixing phenomena for these solutes.

In general, seasonal patterns of concentration for major solutes in the Apure coincided with those of the Orinoco and thus reinforced trends in the Orinoco. The pattern for Si was exceptional, however; it varied little over time in the Orinoco, yet showed a strong seasonal pattern (purging) in the

Apure. Data for soluble Si fit the hyperbolic equation, but violated the model in a geochemical context. Because soluble Si is derived from weathering, concentrations of Si should have been highest during low flow conditions; instead, concentrations were highest during high flow. It has been known for some time, however, that the relationship between Si and discharge is not simple (e.g., Kennedy 1971). A variety of mechanisms regulate Si; their relative importance may even vary spatially within a watershed. The strong purging effect that is characteristic of Si in the Apure is unusual, but not unique (Lesack et al. 1984).

Two mechanisms, soil solute release and biological uptake, should be considered as possible explanations of the inverse relationship between discharge and Si concentration. During the period of high water, a large area of floodplain is inundated by the river. For several months, shallow water covers the floodplain and is thus in close contact with soils. Kennedy (1971) has shown that silica is released very rapidly by soils. If these soils, which are exposed to leaching only during high water, have relatively high Si concentrations, they might augment Si in the river during this hydrological phase and thus explain the purging relationship between concentration and discharge. The absence of a comparable purging effect for other solutes argues against this mechanism, however.

As an alternative hypothesis, uptake of Si by diatoms might deplete soluble Si at times of high diatom growth (Edwards 1973). Diatoms, especially *Melosira granulata*, were the dominant algae in the Apure River (Lewis et al. 1989). Algal biomass and primary production were highest in the Apure at lowest discharge (Lewis 1988). Given primary production at low water of $200 \text{ mg C} \cdot \text{m}^{-2} \cdot \text{d}^{-1}$, most of which was accounted for by diatoms (Lewis 1988), and a travel time of at least four weeks (Saunders & Lewis 1988b), an estimate can be made of biological Si immobilization. Frustules account for approximately 40% of the dry mass of diatoms (Bailey-Watts et al. 1973; Reynolds 1976). Given a mean channel depth of about 2 m, this would indicate uptake of approximately 2 mg Si/L as the river crossed the Llanos. Biological uptake thus provides a plausible mechanism by which Si concentrations could have been reduced at low water in the Apure; maximum rates of uptake closely reflected the seasonal amplitude of variation in Si concentration. This mechanism would result in a purging relationship between concentration and discharge.

The Apure makes a large contribution to the transport of inorganic solutes from the Orinoco watershed, but chemical erosion rates for the Apure basin are not especially high when viewed from a global perspective. On the basis of selected morphoclimatic variables, the Apure River is readily placed in Meybeck's (1979) moist tropical category. In general, the Apure

Table 5. Concentrations and transport of major ions in the Apure River. Moist tropical and global values are from Meybeck (1979).

	Na	K	Ca	Mg	Cl	SO ₄	HCO ₃	SiO ₂	Total
<i>Concentration, mg/L</i>									
Apure	4.10	2.16	16.24	2.99	1.65	10.71	61.78	11.31	110.9
Moist tropical	4.90	1.65	8.25	3.50	4.20	4.00	45.40	11.40	83.3
Global	5.15	1.30	13.40	3.35	5.75	8.25	52.00	10.40	99.6
<i>Transport, tons/km²/yr</i>									
Apure	2.03	1.07	8.02	1.48	0.82	5.29	30.53	5.59	54.8
Moist tropical	1.76	0.59	2.97	1.26	1.51	1.44	16.32	4.10	29.9
Global	1.93	0.49	5.02	1.25	2.15	3.09	19.47	3.89	37.3

carries somewhat higher concentrations of the major solutes than would be expected for rivers of this type (Table 5); concentrations of Ca and SO₄ in the Apure are higher by a factor of 2 or more than those in Meybeck's moist tropical river group. Chloride concentration in the Apure is unusual among the major inorganic solutes because it is lower by a factor of 2.5 than that characteristic of Meybeck's moist tropical river group. Although some rivers in that group may have geological sources of Cl, these are probably insignificant in comparison with atmospheric sources. Concentrations and specific transport rates for the Apure are more closely in line with global than with tropical averages except for Cl, which was conspicuously low; Cl transport is comparable to that of tropical regions receiving much less rainfall. Specific transport of Cl in the Apure is even lower than that of the Caura ($1.03 \text{ t} \cdot \text{km}^{-2} \cdot \text{yr}^{-1}$) where 99% of Cl is derived from atmospheric sources (Lewis et al. 1987). Low transport of chloride is probably the result of the great distance that oceanic air masses must travel before depositing rainfall on the Apure watershed (cf. Meybeck 1983; Stallard & Edmond 1981). The contributions of the Apure watershed to dissolved solids in the Orinoco appear large only because specific transport rates from many other parts of the Orinoco basin are very low (Lewis & Saunders 1988).

Concentrations of most solutes in the Apure River can be modelled as if the watershed were a simple, two-compartment mixing system. The hyperbolic mixing model provided a useful tool for detecting the influences of biogeochemical phenomena other than mixing, and the ability to discern departures from simple mixing was probably enhanced by the use of uniform β . Yields of most solutes from this tropical savanna watershed are comparable to global averages. Nevertheless, the influence of the Apure on transport in the Orinoco is disproportionately large, because yields from the rest of the Orinoco watershed are quite low.

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