

GEOCHEMICAL ASSESSMENT OF ATMOSPHERIC DEPOSITION INCLUDING HARMATTAN DUST IN CONTINENTAL WEST AFRICA

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ABSTRACT In West Africa, atmospheric dust, which is composed of Saharian particles and is transported by Harmattan wind, is a significant seasonal climatic event. The dust primarily was composed of silt-sized particles containing, on average, 75% silica. Variations in the calcium content in the atmospheric deposition provided a means of tracing sources of the deposition. Phosphate was the only constituent measured that varied seasonally. The atmospheric dust affected the chemical quality of rainfall which typically were alkaline and strongly mineralized. A chemical fluxes associated with both wet and dry atmospheric deposition were estimated for continental West Africa. A geochemical assessment suggested the existence of an African chemical signature of the atmospheric fluxes which contribute a mass of about $1300 \text{ kg ha}^{-1} \text{ year}^{-1}$ in Sahelian zone to $365 \text{ kg ha}^{-1} \text{ year}^{-1}$ in Guinean zone. Si was about 33% of the total atmospheric deposition. In contrast, the other chemical species varied spatially and with the type of atmospheric deposition. Over the entire continental West Africa, the relative importance of elements in the dry plus wet atmospheric deposition was as follows: $\text{Si} \gg \text{HCO}_3 > \text{Ca} > \text{Al} > \text{Fe} > \text{Cl} > \text{K} > \text{N} > \text{Mg} (>> \text{Na} > \text{Ti} > \text{S} > \text{P} > \text{Mn})$.

INTRODUCTION

During the last decade, many studies have shown the importance of atmospheric dust transport on African continent and its influence on climate (D'Almeida, 1986, 1989; Tsoar & Pye, 1987; Bergametti *et al.*, 1989; Joussaume, 1990; Legrand, 1990). The present article concerns the role of atmospheric dust on the chemical properties of rainfall in continental West Africa and an evaluation of a geochemical balance of atmospheric deposition, wet and dry. The characteristics and constituent composition of the Harmattan dust falling in West Africa were evaluated then they were compared to those of the rainfall. A chemical assessment was made of the wet and dry atmospheric inputs in continental West Africa.

Daily measurements of dust deposition were conducted at a few sites in Senegal from 1984 to 1989 by ORSTOM. In West Africa, atmospheric dust is as important seasonally as the rainy season (Orange & Gac, 1990; Orange, 1992). Dust deposition ranges from about $2000 \text{ kg ha}^{-2} \text{ year}^{-1}$ in the Sahelian zone and about $400 \text{ kg ha}^{-2} \text{ year}^{-1}$ in the Guinean zone, of which at least 40% of the dust is derived locally from resuspension. The contribution of dustfall to basin sediment budget is negligible and does not change the mechanical erosion balance. But the dustfall may impact the chemical weathering budget by reacting with the rainfall. And the dust deposition is significant during the wet season contributing 25% to the total annual dust flux.

MATERIALS AND METHODS

Dry atmospheric deposition (dust) collectors were installed at three sites in Senegal (Fig. 1). The collector is a pyramidal receptacle 40 cm deep and has a 0.25 cm² collection area (Orange *et al.*, 1990). The top of the collector is located approximately 5 m above the ground and the daily dry deposition to the collector is washed daily with distilled water into a collecting bottle. The samples are filtered through a 0.45 µm filters, dried at 70°C and weighed.

For the chemical analysis of the dry deposition, the samples were composited monthly, which provided: 42 samples for Dakar from 1984 to 1987; 20 samples for Mbour, 80 km south of the Atlantic coast, from 1986 to 1987; and 14 samples for Pete, 400 km east in the continental Sahelian zone, from 1986 to 1987.

Rainfall was collected at two sites in Senegal; one site at Bakel in the Sahelian zone was sampled during the 1983 rainy season and the other site at Kedougou in the Sudanian zone was sampled during the 1987 rainy season (Fig. 1). The rainfall sampling method and some results of chemical analysis are reported by Orange & Gac (1990).

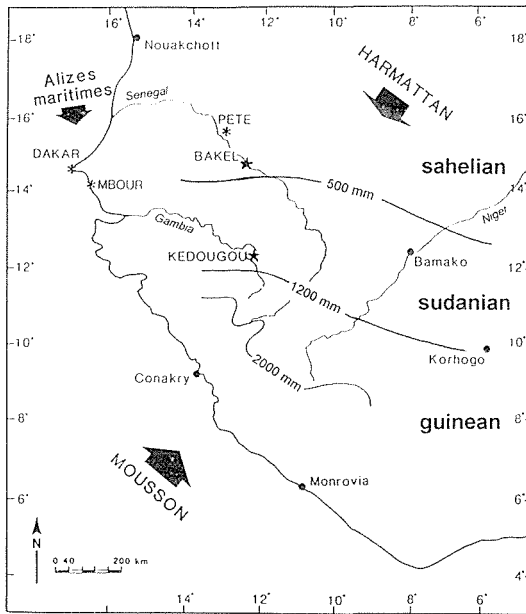


FIG. 1 Geographical location of the sampling area.

COMPOSITION OF HARMATTAN DUST

The dry deposition was primarily composed of silt-sized particles, referred to as the Harmattan dust. Results of an analysis of the size distribution of atmospheric dust at Dakar indicated that 91% of particles ranged from 2 to 50 µm in diameter, 6.5% of them were <2 µm, and only 2.5% were sand size.

The annual mean element deposition was calculated by weighting the monthly analysis results by the monthly average of daily dust deposition. The inter-annual mean was averaging these values (Table 1).

TABLE 1 Annual mean chemical composition, in oxide weight percent, of atmospheric dust collected in Senegal.

Year	DAKAR					MBOUR			PETE		
	1984	1985	1986	1987	1984-1987	1986	1987	1986-1987	1986	1987	1986-1987
n	9	12	12	9	42	12	8	20	9	5	14
SiO ₂	74.3	72.0	74.2	74.7	73.8±2.8	75.9	77.2	76.5±2.6	76.3	77.3	76.8±2.0
Al ₂ O ₃	12.6	12.9	11.3	9.5	11.6±1.9	10.5	9.6	10.1±1.5	10.8	9.8	10.3±0.9
Fe ₂ O ₃	5.1	5.1	5.7	5.7	5.4±0.8	5.4	5.0	5.2±0.6	5.3	5.0	5.2±0.4
TiO ₂	0.95	0.92	0.89	0.82	0.89±0.09	0.88	0.86	0.87±0.10	0.94	0.90	0.92±0.07
MnO ₂	0.073	0.067	0.080	0.084	0.076±0.007	0.100	0.086	0.093±0.009	0.103	0.086	0.094±0.013
P ₂ O ₅	0.224	0.215	0.262	0.276	0.244±0.121	0.248	0.191	0.219±0.076	0.143	0.143	0.143±0.025
CaO	2.04	2.07	2.08	2.46	2.16±0.46	0.97	1.09	1.03±0.30	0.67	1.15	0.91±0.43
MgO	1.47	1.50	1.53	1.54	1.51±0.16	1.51	1.39	1.45±0.21	1.34	1.31	1.33±0.19
K ₂ O	2.20	2.21	1.57	1.01	1.75±0.59	1.05	1.02	1.04±0.12	1.16	1.00	1.08±0.14
Na ₂ O	1.03	1.10	0.76	0.21	0.77±0.42	0.14	0.12	0.13±0.03	0.10	0.11	0.11±0.02

n is number of samples; M±SD where M is the interannual mean and SD is the standard deviation.

For each site, the annual means of dust chemical composition remained relatively constant. Consequently, the inter-annual means provide a good estimate of the characteristics. Furthermore, the composition did not vary markedly among sites and the dust at the sites was dominated by silica (Si). The Si was in quartz (60%) and clay minerals (15-20%) (Orange, 1992). The most abundant oxide after SiO₂ was always Al₂O₃ (10%) and Fe₂O₃ (5%).

Compared to the chemical composition of the soil in this geographic zone (Moberg *et al.*, 1991), atmospheric dust is more siliceous, less aluminous and has about the same ferrous content. Furthermore, atmospheric dust is enriched in Ca, Mg, K, and Na. Consequently, the dust was a source of cations to the typically cation-depleted West African soils. The average chemical composition of the dry deposition, mainly dust, collected at Dakar, Mbour and Pete are listed in Table 2.

The chemical composition of dry deposition did not vary substantially among Dakar, a populated coastal city, Mbour, a low population coastal city, and Pete, a low population continental city. At Dakar, Si was less abundant. The results of the X-ray diffraction analysis indicated that the Si was bound in quartz. At Dakar, the oxides of Ca, K, and Na were more abundant than at the other sites, whereas Mg was similar. The abundance of Ca, K, and Na were probably caused by pollution from this populated area. The dry deposition at Pete contained low concentrations of P compared to the other sampling sites. The high P at the coastal sites may indicate that P can be used as a tracer of dry deposition.

The chemical composition of dry deposition at each site, in general, did not vary seasonally. At Dakar, Si contents were lowest during the wet season, but this did not occur at the other sites. The only chemical constituent that varied annually was P. High P concentrations occurred during wet season and were associated with the burning of organic matter, which is consistent with the hypothesis of Stallard & Edmond (1981) that emissions as particulate from vegetation are the major sources of phosphorus to the atmosphere.

The most variable constituents monthly were, in decreasing order, Na₂O, P₂O₅, K₂O, and CaO at Dakar, P₂O₅, CaO, and Na₂O at Mbour and CaO at Pete (Table 3). Calcium was

TABLE 2 Chemical composition, in oxide weight %, of atmospheric dust in West Africa and a well-drained soil of Northern Nigeria).

Oxide	Dust	Soil (1)
SiO ₂	75.7	49.7
Al ₂ O ₃	10.7	29.6
Fe ₂ O ₃	5.2	4.1
TiO ₂	0.90	1.64
MnO ₂	0.088	0.02
P ₂ O ₅	0.202	-
CaO	1.37	0.08
MgO	1.43	0.44
K ₂ O	1.29	0.90
Na ₂ O	0.34	0.03

(1) Moberg *et al.*, 1991.

TABLE 3 Variations in the major ion composition, in % standard deviation, of atmospheric dust collected at the sites in Senegal.

Oxide	Dakar	Mbour	Pete
SiO ₂	4	3	3
Al ₂ O ₃	17	14	8
Fe ₂ O ₃	14	12	8
TiO ₂	10	12	8
MnO ₂	9	10	14
P ₂ O ₅	50	35	17
CaO	21	29	48
MgO	10	14	14
K ₂ O	34	12	13
Na ₂ O	54	23	17

the only constituent that was highly variable at each site. The variability is attributed to contributions of calcareous crusts during the generation of atmospheric dust (Paquet *et al.*, 1984; Loÿe-Pilot *et al.*, 1986; Coudé-Gaussen, 1989; Clarke & Karani, 1992). These results suggest that Ca can be used as tracer of source regions.

We have no explanation for the high variability of Na and K in the dry deposition, but because their concentrations generally were quite low, some variability may have been associated with analytical precision. Also, no explanation was provided for the assessment of dust in other studies such as Moberg *et al.* (1991).

CHEMICAL PROPERTIES OF RAINFALL

Compared with world average rainfall, the continental Sahelo-Sudano-Guinean rainfall was highly mineralized ranging from 10 mg l⁻¹ to 50 mg l⁻¹, and was dominated by HCO₃ and Ca (Mathieu, 1976; Roose, 1980; Lewis, 1981; Travi *et al.*, 1987; Orange & Gac, 1990; Yaïr *et al.*, 1991). Orange & Gac (1990) showed that dissolved solute concentrations increased with increasing aridity. The concentrations were enhanced when monsoon cloud masses merge with the Harmattan winds which are charged with dust particles. Furthermore, the pH of the rainfall was slightly alkaline. Rainfall acidity, which is noted in tropical forests south of this area (Loÿe-Pilot *et al.*, 1986; Jaffrezo, 1987; Lacaux *et al.*, 1987; Caboi *et al.*, 1992), is counteracted by the interaction of rainfall with the calcareous Harmattan dust.

At Bakel, the highest solute concentrations in rainfall were at the beginning and the end of the wet season; solute concentrations were lowest wet season (Table 4). Consequently, rainfall played a significant role in the cleansing of the atmosphere. This phenomenon has been reported in other studies (Buat-Ménard *et al.*, 1974; Lewis, 1981; Jaffrezo, 1987; Orange & Gac, 1990).

TABLE 4 Monthly chemical characteristics of rainfall collected at Bakel, where: solute concentrations are in $\mu\text{eq l}^{-1}$; SiO_2 is in $\mu\text{mol l}^{-1}$; Pp is the collected amount of rain water, in mm; Npp is the number of storms; and TDS is the total dissolved solids concentration, in mg l^{-1} .

Parameter	June	July	August	September	October
Pp	73	25	172	38	12
Npp	4	1	9	5	3
HCO_3	415	123	80	75	235
Cl	118	63	61	84	122
SO_4	40	64	28	44	66
NO_3	35	12	9	19	31
PO_4	11	3	1	10	14
Ca	246	98	57	103	218
Mg	194	22	15	26	46
K	78	12	8	16	40
Na	149	15	16	34	49
NH_4	1	110	70	46	109
SiO_2	40	4	3	8	20
TDS	52.7	18.9	12.4	16.2	35.1

For dissolved Si in rainfall, suspended soil particles, referred to as atmospheric dust, is the only source. For the other elements, the load increases caused by the dissolution and washout of the dust in the lower atmosphere was computed by taking the ratio of the element concentration in rainfall to Cl concentration in rainfall and comparing that ratio with one for seawater as follows:

$$E_d(X) = [(C_X/C_{Cl})_{\text{rainfall}} / (C_X/C_{Cl})_{\text{ocean}}] - 1 \quad (2)$$

where $E_d(X)$ is the enrichment factor for element X, C_X is the concentration of the element X, and C_{Cl} is the concentration of Cl, used as reference. The Cl concentration characterizes the marine source, which is consistent with previous studies (Stallard & Edmond, 1981; Meybeck, 1984). If $E_d(X)$ is positive, then the element X is enriched in rainfall. The oceanic ratios used for comparison, $(C_X/C_{Cl})_{\text{ocean}}$, were those reported by Savenko (1976).

The element enrichment results are listed in Table 5. The results indicate that elements had the highest enriched in the rainfall of June and October. Magnesium was enriched only in June whereas HCO_3 , Ca, and K were enriched during the entire wet season. Bicarbonate was the most enriched of all elements and can again be attributed to the rainfall interaction with calcareous crusts on the dust particles.

GEOCHEMICAL BALANCE OF ATMOSPHERIC DEPOSITIONS

The uniformity of chemical composition of West African rainfall and atmospheric dust, as noted by Orange & Gac (1990), allows us to calculate a geochemical flux associated with atmospheric deposition by using data collected in the three climatic zones: Sahelian, Sudanian and Guinean zones.

The average chemical composition of dissolved constituents in atmospheric deposition of continental West Africa was derived from the volume-weighted chemical concentrations of rainfall collected at Bakel in Sahelian zone, at Kedougou in Sudanian zone, and at

Korhogo, Ivory Coast, in Guinean zone (Roose, 1980). The volume-weighted concentrations for these areas are listed in Table 6.

TABLE 5 Enrichment factors, $E_d(X)$, of chemical major constituents (X) in rainfall collected at Bakel.

X	June	July	August	September	October
HCO ₃	151	84	56	38	83
SO ₄	0	2	0.3	0.5	0.5
Ca	15	11	6	8	12
Mg	7	0.6	0.2	0.5	0.8
K	20	5	3	5	9
Na	0.4	-0.7	-0.7	-0.6	-0.6

$$E_d = [(X/Cl)_{rain} / (X/Cl)_{ocean}] - 1$$

The average volume-weighted, total dissolved solutes concentration of the rainfall in continental West Africa is about 13 mg l⁻¹. In terms of equivalent of charge, the relative importance of the soluble cations was Ca>NH₄>Mg>Na>K. For soluble anions, the relative importance was HCO₃>Cl>SO₄>NO₃>PO₄. The dissolved Si concentration was low but much higher than that of the world average. The dominant major ion was HCO₃ (Fig. 2). Lewis (1981) reported the same order of importance for the soluble anions in a tropical watershed of Venezuela which is in a similar environment. For the soluble cations, the relative contribution of Ca and NH₄ was more important in our study than in Venezuela, largely because of the high contents of Harmattan dust in continental West African atmospheric deposition.

TABLE 6 Average chemical composition, in mg l⁻¹ except SiO₂ in μmol l⁻¹, of dissolved constituents in atmospheric deposition in Sahelian, Sudanian and Guinean climatic zones.

Constituent	Sahelian Zone	Sudanian Zone	Guinean Zone	Continental West Africa
HCO ₃	9.5	6.5	4.8	6.2
Cl	2.7	1.2	0.3	1.0
SO ₄	2.0	1.0	1.0	1.1
NO ₃	1.0	0.5	0.5	0.6
PO ₄	0.2	0.3	0.3	0.3
Ca	2.2	1.5	1.9	1.8
Mg	0.6	0.2	0.1	0.2
K	0.9	0.6	0.2	0.5
Na	1.0	0.3	0.3	0.4
NH ₄	1.2	1.0	0.3	0.7
SiO ₂	0.7	0.4	0.7	0.6
TDS	22.0	13.5	10.4	13.4

TDS, Total Dissolved Solute in mg l⁻¹.

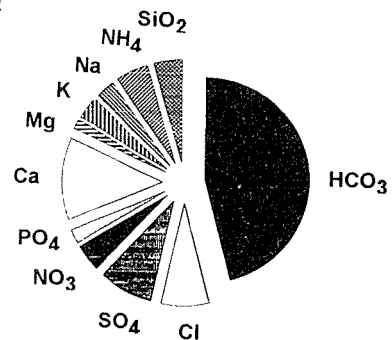


FIG. 2 Chemical characteristics of rainfall in continental West Africa.

The geochemical balances of atmospheric deposition was calculated by using total rainfall quantities from a hydroclimatic study of West Africa by Orange (1992). The average annual rainfall was 400 mm in Sahelian zone, 1100 mm in Sudanian zone, and 1200 mm in Guinean zone. Assuming a resuspension rate of 40% (Orange, 1992), the mean annual deposition of atmospheric dust was 120 g m^{-2} in Sahelian zone, 70 g m^{-2} in Sudanian zone, and 25 g m^{-2} in Guinean zone.

The flux estimate indicates that atmospheric dust dominates the element deposition in West Africa, regardless of the climatic zones (Fig. 3). The dust contributed 93% of the total mass deposition to the Sahelian zone, 83% to the Sudanian zone, and 66% to the Guinean zone. In continental West Africa, the flux of atmospheric dust contributed from 2 and 14 times the element mass flux of that in rainfall.

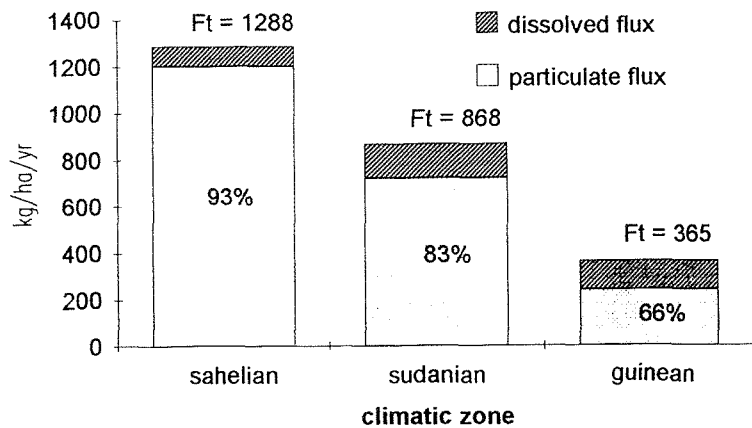


FIG. 3 Dry and wet atmospheric deposition in continental West Africa where: F_d is the dry deposition, in $\text{kg ha}^{-1} \text{ year}^{-1}$; F_p is the wet deposition, in $\text{kg ha}^{-1} \text{ year}^{-1}$; F_t , total flux of atmospheric inputs; contribution of dust deposition, in %.

Atmospheric deposition was highest in the Sahelian zone, because of the large contribution of dust particles to the total in this climatic zone. It was about $1288 \text{ kg ha}^{-1} \text{ year}^{-1}$ in the Sahelian zone and was only about $365 \text{ kg ha}^{-1} \text{ year}^{-1}$ in the Guinean zone; that represents a variation of 72% between these two extremes. In the Sudanian zone, the total atmospheric deposition was $868 \text{ kg ha}^{-1} \text{ year}^{-1}$.

The fluxes of individual constituents in rainfall and in dry deposition (dust) are listed in Table 7. Rainfall contributes all of the carbon, as HCO_3 , Cl, N, and S. The fluxes of P, Na, Ca, K, Mg, and Si varied in the two forms depending on the climatic zone. For example, dry deposition dominated the Ca atmospheric flux in the Sahelian zone whereas rainfall dominated the flux in the Guinean zone. Si was always present and dominated by dry deposition. Finally, Al, Fe, Ti, and Mn were exclusively deposited by dry deposition.

The form of atmospheric deposition and relative contribution of chemical species varied across the climatic zones (Table 7). However, the relative importance of the atmospheric inputs of chemical species was similar among the climatic zones and each was dominated by Si, HCO_3 , Ca, Al, and Fe. Si was always the most abundant chemical element deposited from the atmosphere onto the continental West African landscape and contributed about 33% to the total mass input. Following Si, HCO_3 flux was the second

largest and varied from 3% of the total flux in the Sahelian zone to 16% in the Guinean zone. Calcium was more or less abundant than Al or Fe, depending on the relative importance of dry deposition to the total flux for a zone. In the Sahelian zone, Al and Fe fluxes were greater than that for Ca, whereas in the Sudanian and Guinean zones Ca was more important (Table 7). For the other elements, the order of importance was not consistent among zones.

TABLE 7 Chemical composition of atmospheric deposition in continental West Africa, in $\text{kg ha}^{-1} \text{ year}^{-1}$.

Deposition Type	Constituent	Sahelian Zone		Sudanian Zone		Guinean Zone		Continental West Africa
		<i>Fd</i>	<i>Fp</i>	<i>Fd</i>	<i>Fp</i>	<i>Fd</i>	<i>Fp</i>	<i>Ftotal</i>
wet only	HCO ₃	38.0		71.5		57.6		71.5
	Cl	10.8		13.2		3.6		13.2
	N	4.6		9.8		4.2		9.8
	S	2.7		3.7		4.0		3.7

wet and dry	P	0.3	0.5	1.1	0.3	1.2	0.1	1.4
	Na	4.0	1.5	3.3	0.9	3.6	0.3	4.2
	Ca	8.8	11.7	16.5	7.0	22.8	2.3	23.5
	K	3.6	6.4	6.6	3.8	2.4	1.3	10.4
	Mg	2.4	10.3	2.2	6.2	1.2	2.1	8.4
	Si	1.3	424.8	2.1	254.9	3.9	85.0	257.0

dry only	Al		33.8		20.3		6.8	20.3
	Fe		22.0		13.2		4.4	13.2
	Ti		6.4		3.9		1.3	3.9
	Mn		0.7		0.4		0.1	0.4

Fd, dissolved flux; *Fp*, particulate flux.

Finally, estimates of the mass contribution of each element or constituent in wet and dry atmospheric deposition to continental West Africa are listed in Table 7. The fluxes to the Sudanian zone were taken to be those for continental West Africa. The annual total atmospheric deposition to continental West Africa was $850 \pm 50 \text{ kg ha}^{-1} \text{ year}^{-1}$, of which 33% was Si. The chemical mass balance of dry and wet atmospheric inputs in continental West Africa is in the following order: Si > HCO₃ > Ca > Al > Fe > Cl > K > N > Mg (> Na > Ti > S > P > Mn).

CONCLUSIONS

The geochemical balance of atmospheric deposition underscores the importance of Harmattan dust in continental West Africa. Wet precipitation was a significant process in the cleansing of the atmosphere, and the water soluble fraction of the tropospheric aerosols governs the dissolved salt content of rainfall.

The chemical composition of dry atmospheric deposition in Senegal which primarily

consisted of dust, was relatively constant over the entire study area. The dust was dominated by silt-sized particles of which Si was about 75% of the total mass deposited. Also, dust was a source of cations for the soil formation. The chemical composition did not vary seasonally with the exception of P which had higher contributions during the wet season when vegetation typically is burned. Calcium concentrations and deposition were highly variable and may prove useful as a tracer of the dust.

The dry atmospheric deposition contributes significantly to the total deposition and has some major geochemical implications because it not only contributes elements by dry deposition, but it also is washout of the atmosphere by rainfall and interacts with it. The enrichment, particularly HCO_3 , Ca, K, and Si, was highest at the beginning and end of the wet season due to the flushing of the dust particles in the lower atmosphere. The constituent most enriched was HCO_3 , a result of the reaction of rainfall with calcareous dust.

Atmospheric dust was always the major part of atmospheric inputs regardless of the climatic zones. The dry atmospheric deposition was the highest in the Sahelian zone and the deposition varied by 72% between the two extremes. The deposition of P, Na, Ca, K, Mg, and Si varied markedly between the two forms of input among the zones. Si was always present particularly in the dry deposition. The relative importance of the element deposition was similar among the climatic zone and was dominated by Si, HCO_3 , Ca, Al, and Fe. The annual total atmospheric deposition to continental West Africa was $850 \pm 50 \text{ kg ha}^{-1} \text{ year}^{-1}$, of which 33% was Si.

These results underscore the importance of atmospheric dust to the geochemical fluxes and the chemistry of waters, particularly rainfall. Knowledge of the geochemistry of Harmattan dust is necessary for understanding chemical weathering of West Africa landscapes.

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