Impact of termite activity on soil environment: A perspective from their soluble chemical components

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ABSTRACT: An investigation on varied types of termite mounds relative to the nearby soils that are not inhabited by the termites in different places of Cameroon show that the activity of the termites is increasing the contents of most major and some trace elements in the termite mounds, except for Si and sometimes Fe, Mn, Na and K. These released elements are relocated into newly formed mineral phases that are dissolved by either H_2O or dilute HCl leachings. The Ca and Mn released by the termite activity testify for crystallization of Ca-Mg carbonates and phosphates as well as of Fe oxy-hydroxides and/or Mn hydroxides. Termite activity also induces an increase in the lanthanide contents, the mound materials being especially enriched in light lanthanides relative to the corresponding soils without termite activity. The shapes of the patterns support precipitation of Mn-Fe oxy-hydroxides and Ca carbonates-phosphates. The increased amounts of Eu and Ce linked to termite activity seem to relate to the occurrence of reducing agents that are released by the termites, modifying Eu^{+3} into Eu^{+2} and Ce^{+4} into Ce^{+3}, favoring in turn selective incorporation of Eu^{+2} and Ce^{3+} in the new phases of the termite mounds. Another consequence of the termite activity is the precipitation of H_2O and HCl extractable phases having low Sr/Ca ratios. Even if the K/Rb values of the termite mounds are typical for common soil-forming silicate minerals, their relocation by an inorganic process alone does not explain an abnormally high ratio in the H_2O leachable mineral phases. It was also shown that the main source for K and Rb of the dissolved phases is not only the interlayer site of clay particles, but also nutrients immobilized in and by the termites.

Key words: *Trace elements, lanthanides, weathering, crystallization, ground and surface waters, termite activity*

INTRODUCTION

Termites are common biological agents that produce significant physical and chemical modifications to tropical and subtropical soils (Nye, 1955; Sys, 1955; Pomeroy, 1976; Briese, 1982; Akamigbo, 1984; Lobry de Bruyn and Conacher, 1990 and 1995; Mando *et al.*, 1996; Heikens *et al.*, 2001). They generally go through a sequence of actions, from fetching, carrying to cementing mineral particles into mounds by using their salivary secretion (Howse, 1970; Wood, 1988; Donovan *et al.*, 2001; Ndiaye *et al.*, 2004; Vandecasteele *et al.*, 2004; Lopez-Hernandez *et al.*, 2006). For example, it has been shown that termite activity increases the content of organic matter in the soils that they use for the construction of their nests and also modifies the clay mineral composition of these soil materials (Mahaney *et al.*, 1999; Jouquet *et al.*, 2002; Roose Amsaleg *et al.*, 2004). Abundant literature can be found related to effects of termites on the mobility of a number of soil elements, but the focus is largely on those few elements that are generally considered to be essential for the support or growth of all forms of life (Jouquet *et al.*, 2004; Brossard *et al.*, 2007). The same literature has shown hardly any interest in examining the effect of termite activity on the mobility of trace elements that are generally considered to be non-essential to living systems. Nevertheless, it can not be altogether denied that the dynamics of mobility of trace elements may hold significant clues as to understandings of the processes of reorganization of soil minerals and relocation of chemical elements that

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constitute the frameworks of soil minerals. This report has made an attempt to depict characteristics of geochemical changes among several minor or trace elements, such as Rb, Sr, Th, U, Ar and the lanthanide group, in conjunction with major chemical and mineralogical changes, that may be produced in soil materials through termite activity. The lanthanide group consists in 14 elements ranging from La with atomic number 57 to Lu with atomic number 71. Lanthanides and rare earth elements (REE) are synonymous words, the former term being used frequently by biologists and soil scientists, where the latter is used more popularly by geologists and geochemists.

It might be added that the results of this study cover only a limited geographical area with narrowly defined termite species and can not be applied universally to geochemical modifications produced in soils by termite activity, because termite species are widely varied with a wide range of living, breeding, and feeding habits.

However, it might be of interest to note that (1) termites generally built epigeous nests having homogenous structures, even if the nests have varied shapes, (2) the materials needed for the construction of the nests are taken by the termites from immediate environment, generally limited to a few meters surrounding the nested area, (3) the mineral particles of the soils represent often the basic used material, and (4) termites mix soil materials with wood and excrements for the construction of their nests (Noirot, 1970; Jungerius *et al.*, 1999). In fact, the supply of fine clay particles is adequate for building the nests or mounds that are characterized by thick, dense and solid outer layers, protecting chambers and communicating passages of the mounds. It must be of interest that sponge-like mass of chewed wood fills most of the chambers together with fungus-like material. Because termite activity could have the potential of causing the development of newly-formed materials, this report also provides an account of the amorphous materials that may be derived from their activity.

MATERIALS AND METHODS
Materials
Four pairs of samples were collected in different localities of Cameroon (Ndma Ngoupayou, 1997) that are representative of varied environments in either natural sites away from any human activity, or in places where the natural environment was replaced by human agricultural activity. Each pair of samples represents a termite-affected material taken from a mound/nest (labelled conventionally termite "mound") and the corresponding soil sample taken several meters away from closest mound and considered to be the reference without significant termite activity (labelled conventionally "soil" without termites), not meaning necessarily that no termite activity never occurred in it. The sample pair 1 comes from southern Cameroon near a village called Nsimi; the mound being constructed on a tree stock, consisting in indurated, laminated material in a forested area, with a nearby sand-clay type soil at 3 m. from mound. The sample pair 2 was collected next to a village named Dibamba, close to Douala; the mound being also constructed on a tree stock in a humid region close to the ocean were the original forest vegetation was replaced by a traditional agriculture without fertilizers on a yellowish sand. The sample pair 3 was taken at Yambassa near Ebebda in central Cameroon in an open grassy, savanna-type, region on a sandy clay rich soil. The sample pair 4 comes from Fomopea near Dschang in western Cameroon; the mound being constructed on a grassy slope at an altitude of 1500 m. on a soil deriving from clay-rich sedimentary rocks.

Methods
The termite mounds were excavated and dismantled. Samples of about 2 kg which were thoroughly mixed by repeated (more than 10 times) conning and quartering. They were dried for 5 days at room temperature, and placed afterwards in an oven at 60 °C for about 10 h. before being crushed in a mortar and split into five aliquots. One fraction was analysed for the mineral composition by X-Ray diffraction (XRD). A second aliquot was first leached with double distilled, deionized H_2O during 15 min. for chemical analysis of the leachate, followed by a second leaching with 1N HCl again for 15 min. at room temperature also for chemical analysis of the leachate. Another aliquot was used for extraction of the amorphous phases, and the remaining powder was taken for both major and trace element analysis and for K-Ar isotopic tracing.

Extraction of the amorphous phases
The amorphous phases of the termite mounds and soils without termites were extracted as following methods of Chao and Zhou (1983). The samples were first treated with Morgan's reagent, which consists of a 1M sodium acetate solution at pH 5, to remove the

exchangeable ions from mineral and organic components. Then, the Tamm reagent consisting of 16.1 g/L ammonium oxalate and 10.9 g/L of oxalic acid, was used to dissolve the amorphous components. The procedure consisted in a mixing of 40 mL of Morgan's reagent pro g of soil shaken together for 4 h. at room temperature (25 °C) in a polypropylene tube. Each sample was treated afterwards with 40 mL of Tamm's reagent in the dark at room temperature (25 °C).

Laboratory analyses

Chemical and mineralogical investigations were carried out on the bulk samples and the <2 μm size fractions of the mounds build by the termites and the corresponding soils without termites. The <2 μm fractions were separated from bulk samples of two sites by sedimentation in double-distilled water, followed by a centrifugation to concentrate the separated fractions. The mineralogy of both the bulk samples and the size fractions were analyzed by XRD. One XRD diagram of a disordered powder from bulk sample as well as three XRD diagrams of the < 2 μm oriented smear slides, (1) after drying at room temperature, (2) after glycol solvation and (3) after heating at 490 °C for 4 h. were run.

Chemical analyses were performed on an ICP-AES (ARL 3500 C) for the major elements and some trace elements. Some trace elements were determined on an ICP-MS (VG Plasmaquad PQ2+) following a method described by Samuel *et al.* (1985). The analytical precisions are known to be at 5 % for the major elements and at 10 % for the trace elements (at the 2σ level) by repetitive analysis of standard powders during the course of the study.

RESULTS AND DISCUSSION

The results are presented in successive sections that detail the data of the bulk samples and the <2 μm fractions of the termite mounds and soils without termites, and the H_2O and dilute HCl extracts. The results are then followed by their discussion.

Bulk samples

The termite mounds consisted dominantly of clay minerals with quartz, whereas the corresponding soils without termites contained more quartz than clay minerals. Small amounts of Fe-oxi-hydroxides were present in both soils, but relatively more in the termite mounds.

The XRD analyses indicated that the clay minerals were essentially kaolinite with a minor amount of chlorite. The bulk samples consist mainly in Si, Al and Fe and secondarily in Mg, Ca, Na, K, Ti and Mn (Table 1). A comparison of the materials from mounds 1, 2 and 3 with the corresponding soils without termite activity reveals that termite activity apparently increased concentrations for a number of major elements, such as Al, Ca, Mg, K, and Ti, but markedly decreased the concentrations of Si. Like many of the major elements, the concentrations of Fe and Mn in materials from mounds 1 and 2, but not in material from mound 3, were also found to have been increased relative to that in corresponding soils without termite activity. In mound 3, the Fe concentration apparently remains unchanged and the Mn concentration rather decreases. The termite activity in mound 4 apparently produces no significant change in the bulk chemical compositions, unlike noticeable major chemical changes relative to the other three mounds.

In each case, the Sr/Ca ratio of the material from termite mounds is lower than that of the corresponding soils without termite activity. The Sr/Ca ratio for materials from mounds ranges from 7.5 to 174 x 10^{-3} and that for the corresponding soils without termite activity from 98 to 245 x 10^{-3}, the decreases due to termite activity are because of relatively higher increases in Ca than in Sr (Table 1).

The K/Rb ratio ranges from 127 to 401 for the materials from mounds and that for the corresponding soils without termite activity from 36.5 to 388. The data for the materials from mound 1 and mound 2 relative to the data for the corresponding soils without termite activity suggest that termite activity in these two cases increases the K/Rb ratio, the increases in the K/Rb ratio for each being more due to an increase in the K concentration than an increase in the Rb concentration. The K/Rb ratio appears not to be affected by termite activity in the other two cases, where the ratio in each instance remains unchanged, although both K and Rb concentrations increase for the material from mound 3, whereas they remain unchanged for the material from mound 4, relative to the concentrations of the corresponding soil materials.

The K-Ar isotope analyses indicate that the K-Ar dates for the samples from three termite mounds (1, 3 and 4) to be lower than that for the corresponding soils without termite activity: 652 Ma for the material from mound 1 versus 886 Ma for the corresponding soil

without termite activity, 486 Ma for that from mound 3 versus 615 Ma for the corresponding soil without termite activity, and 368 Ma for that from mound 4 versus 403 Ma for the corresponding soil without termite activity (Table 1). The differentials in the dates were not related to the same factors for all three sets of samples. Termite activity apparently induces decreases in the content of radiogenic ^{40}Ar for the samples of two termite mounds (1 and 4), but an increase in the K content for the material from mound 3 and no change in the K concentration for the material from mound 4, relative to their corresponding samples without termite activity (Table 1). By contrast, the materials related to mound 3 show increases in both ^{40}Ar and K concentrations.

The total lanthanide concentrations (i.e., the sum of concentrations of La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb and Lu) for three of the four mounds, namely mounds 1, 2 and 4, are relatively higher than the corresponding soil samples without termite activity; the sample from the remaining mound 3 has a lower total lanthanide concentration than the corresponding soil without termite activity (Table 1). The total lanthanide concentration ranges from 142 to 329 μg/g for the termite mounds and ranges from 92 to 536 μg/g for the corresponding soils without termite activity.

The post archean Australian shale (PAAS), (McLennan, 1989) normalized relative to the distribution of the lanthanides for each of the mound sample and soil sample without termite activity are characterised by downward concave patterns with enrichment of the middle lanthanides from Eu to Er, but with varied degrees of Ce and Eu anomalies, some being not significant at analytical uncertainty. Considering the PAAS-normalized relative distribution patterns, the sample from mound 1 and the corresponding soil without termite activity have negative Ce anomalies, but no Eu anomaly. The sample from mound 2 has both Ce and Eu positive anomalies, but the corresponding soil sample without termite activity has neither a Ce anomaly, nor an Eu anomaly. The sample from mound 3 outlines only an Eu positive anomaly and the corresponding soil without termite activity has a Ce negative anomaly as well as a positive Eu anomaly. The sample from mound 4 and the corresponding soil sample both have a Eu positive anomaly (Fig. 1a). The distribution patterns of the lanthanides of the samples from the four mounds with the values normalized to the lanthanide values of the corresponding soils without

termite activity are shown in Fig. 1b. This comparative basis provides three distribution patterns for the samples from different mounds. The sample from mound 1 has a light REE enrichment (which can, in another way, be described as a heavy REE depletion) distribution pattern, whereas the samples from mound 2 and mound 4 both have almost flat lanthanide distribution patterns, but with a marked negative Eu anomaly for the sample from mound 2. By contrast, the sample from mound 3 has, broadly speaking, a concave upward lanthanide distribution pattern with a marked negative Gd anomaly.

Amorphous materials

The amounts of amorphous materials in the samples from mounds and in the corresponding soils without termite activity are presented in Table 2. The amounts from mounds range from 0.06 to 10.8 mg/g, whereas those from soils without termite activity range from 0.04 to 10.7 mg/g. Only a significant difference in the amount of amorphous material exists between the material of mound 2 with a value of about 0.5 mg/g and the corresponding soil without termite activity with a value of 0.04 mg/g. This relatively higher value for the mound 2 material is an effect of an increase in Al in conjunction with decrease in Si. The differences in the amounts of amorphous materials for the other three cases were relatively quite small. Nevertheless, there are relative decreases in Si concentrations and increases in Al concentrations for materials from three mounds (namely mound 1, 2 and 3), when compared to the concentrations of the same elements for the corresponding soil materials without termite activity.

Clay fractions

The clay fraction (less than 2 micrometers in diameter) of the material from termite mounds yields higher concentrations of Ca, Sr, Na, K, Fe and P, but with about no changes in Si, Al, Ti and Rb concentrations (Table 3). Termite activity appears to have increased the K/Rb ratio, as the clay fractions of the soils without termite activity have a K/Rb ratio of about 198 in contrast to the clay fraction of the mounds with a ratio of about 282. The Sr/Ca ratio decreases from 57 x 10^{-3} for the clay fraction from the soils without termite activity to 11 x 10^{-3} for the clay fraction of the material from mounds. This decrease results from a greater increase in the Ca concentration than in the Sr concentration. The total lanthanide concentration of

Table 1: Chemical composition of the termite mounds (labelled TM) and the corresponding soils without termites (labelled SWT) in major, trace and rare-earth elements

Major (mg/g)	Sample pair 1		Sample pair 2		Sample pair 3		Sample pair 4	
	TM	SWT	TM	SWT	TM	SWT	TM	SWT
Si	289.1	334.4	194.4	372.5	341.4	387.5	292.4	317.0
Al	143.3	115.3	130.4	66.7	83.9	67.2	119.7	107.2
Mg	3.43	1.08	22.8	0.46	1.95	1.24	3.55	3.00
Ca	9.82	0.24	131.3	0.23	1.46	0.78	0.65	0.55
Fe	51.8	41.9	53.0	26.6	31.7	31.9	61.0	56.5
Mn	0.90	0.18	4.55	0.10	0.76	2.87	0.39	0.32
Ti	9.01	7.92	11.4	10.23	7.18	4.17	7.06	5.91
Na	0.16	0.17	1.41	0.08	1.76	1.37	0.59	0.74
K	3.69	2.16	28.3	0.54	28.6	19.5	20.0	22.3
P	1.11	0.59	17.37	1.66	0.33	0.14	0.49	0.43
Total	512.4	503.9	595.0	479.2	499.0	516.7	505.8	514.0
Sr/Ca (10^{-3})	11.9	97.9	7.51	212.5	92,3	129.4	174.0	245,4
K/Rb	126.7	103.6	223.2	36.49	400.6	388.1	181.1	208.4
Ar (%)	16.9	2.8	ND	ND	83.6	88.6	79.4	85.4
^{40}Ar (10^{-6} cm^3/g)	6.58	16.90	ND	ND	62.62	53.18	32.33	38.96
K-Ar (Ma $\pm$ 2σ)	652 (31)	886 (41)	ND	ND	486 (14)	615 (22)	368 (13)	404 (13)
Trace (µg/g)								
Sr	116.3	23.7	984.9	48.9	134.7	100.9	113.1	135.0
Ba	122.7	83.5	bdl	96.7	2340	1941	756.5	880.7
V	133.2	109.5	189.4	109.7	78.7	83.0	162.8	150.5
Ni	63.4	47.4	bdl	15.2	12.9	9.46	23.8	21.0
Co	19.0	13.5	bdl	bdl	8.62	18.9	13.6	13.3
Cr	152.3	136.6	132.6	91.2	28.0	27.3	40.7	38.7
Zn	80.4	58.7	bdl	31.5	42.0	30.5	64.5	58.6
Cu	50.8	24.8	208.3	10.9	14.0	11.6	29.4	31.0
Zr	526.5	584.6	681.8	693.0	2259	1287	424.1	351.9
Rb	29.1	20.9	126.8	14.8	68.9	48.6	110.4	107.1
Pb	ND	ND	6.01	24.1	34.0	114.1	32.4	32.8
Th	19.6	15.2	27.2	17.9	12.1	7.49	35.9	29.7
U	2.89	2.53	3.67	2.17	2.62	1.69	9.81	8.6
U/Th	0.15	0.17	0.14	0.12	0.22	0.23	0.27	0.29
REE (µg/g)								
La	33.5	23.3	79.4	44.8	41.7	26.6	42.1	35.2
Ce	68.4	41.0	141.0	98.5	120.5	473.3	119.4	109.2
Pr	6.09	4.02	16.5	9.46	7.42	5.21	9.84	8.41
Nd	20.4	13.4	60.8	34.5	26.0	17.7	36.8	31.8
Sm	3.41	2.26	11.0	6.19	3.74	2.71	6.92	5.98
Eu	0.70	0.51	2.31	1.24	1.17	0.92	1.60	1.44
Gd	2.48	1.84	7.32	4.12	3.14	4.02	4.99	4.39
Tb	0.41	0.31	0.96	0.55	0.41	0.34	0.72	0.63
Dy	2.31	1.81	4.48	2.51	2.44	1.65	3.58	3.26
Ho	0.52	0.42	0.84	0.45	0.57	0.38	0.73	0.65
Er	1.38	1.11	2.16	1.22	1.81	1.17	1.97	1.76
Tm	0.23	0.19	0.33	0.20	0.35	0.22	0.31	0.29
Yb	1.55	1.37	2.20	1.34	2.71	1.73	2.10	1.95
Lu	0.25	0.23	0.37	0.23	0.57	0.35	0.36	0.33
Total	141.6	91.6	329.4	205.3	212.5	536.3	231.4	205.3
Ce/Ce*	0.78	0.54	1.98	1.12	0.98	0.64	1.09	0.93
Eu/Eu*	1.13	1.18	1.20	1.15	1.61	1.26	1.28	1.32

Ce* = [3*Ce/Ce$_{(PAAS)}$]/[(2La/La$_{(PAAS)}$) +Nd/Nd$_{(PAAS)}$] and Eu* = [(Sm/Sm$_{(PAAS)}$) + (Gd/Gd$_{(PAAS)}$)]
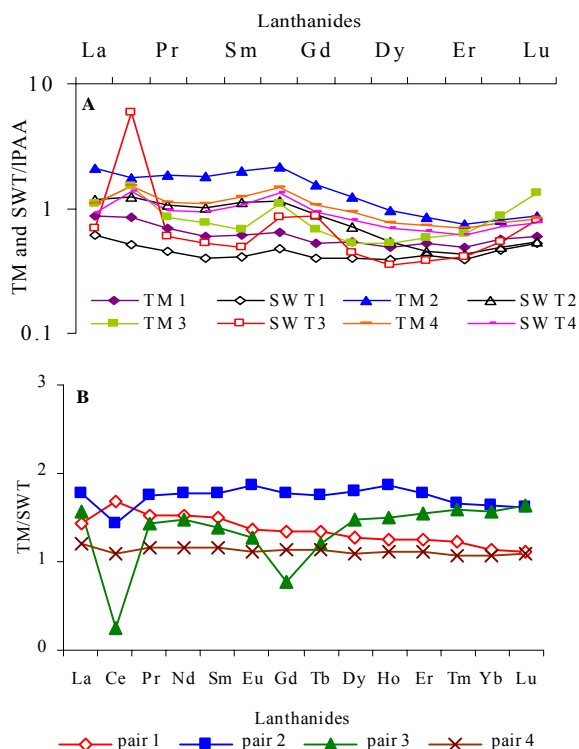bdl = below detection limit; ND = Not Detectable

Fig. 1: Concentrations of the lanthanides in the sample pairs normalised (A) to the lanthanides in the Post Archean Australian Shales (PAAS) and (B) of the lanthanides from termite mounds normalysed to those in the soils without termites. TM stands for termite mounds and SWT for soils without termites
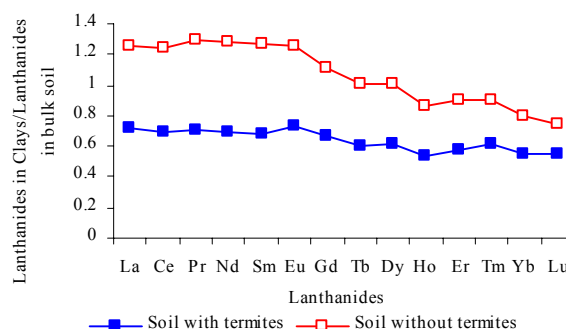

Fig. 2: Concentrations in lanthanides of the <2 μm clay fractions normalized to the concentrations of the corresponding termite mounds and soils without termites called bulks

the clay fraction of the material from mounds is about 99 μg/g, whereas that of the clay fraction from the corresponding soils without termite activity is about 113 μg/g. Within the limits of the analytical error, the two values can be seen as being nearly identical. The distribution of the clay lanthanides from mounds normalized to the lanthanides of the soils without termites may be described by an enrichment in light lanthanides relative to heavy lanthanides with a slight positive Eu anomaly (Fig. 2).

H$_2$O and HCl extracts

As already mentioned, the total dissolved constituents of the H$_2$O extracts represent mainly the elements that were adsorbed on the mineral particles and organic compounds. For the termite mounds, they range from 0.09 to 15.1 mg/g and for the soil without termites from 0.06 to 0.07 mg/g (Table 4). The K and Ca contents collectively contribute to more than 27 % of

the total dissolved constituents of the H$_2$O extracts from termite mounds, and the Si, Al and Mg each contribute about 3 to 30 % of the total value. In contrast, the total dissolved constituents of the H$_2$O extracts from soils without termites are largely dominated by Si, Al and K, collectively representing nearly 23 % of the total dissolved amount in most samples. The H$_2$O extract from termite mounds has a K/Rb ratio of 184 to 1040 in contrast to that of the H$_2$O extracts from soils without termite that varies narrowly from 120 to 161. A measurable difference exists also in the Sr/Ca ratio from H$_2$O extracts: that from mounds is of 4.2 to 11.1 x 10^{-3} and that from soils without termites of 8.6 to 13.5 x 10^{-3}.

The general trend of the major element concentrations in the H$_2$O leachates from all mounds but mound 3 is an increase, while being depleted in Mn, Al, Fe, and Ti, and remaining unchanged in Si concentrations. The H$_2$O leachate of the mound 1 is depleted in Ti and Na and that of the mound 4 is enriched in all elements, except in Mg.

The HCl extracts contain the elements released from soils subsequent to H$_2$O leaching. They represent the components of the mineral and organic phases that were dissolved by the dilute HCl. Those of the termite mounds yield a total dissolved constituent of 0.96 to 133 mg/g of soil, whereas those of the soils without termites have a significantly lower total dissolved constituent of 0.38 to 1.02 mg/g of soil (Table 5). Ca represents more than 30 % of the total dissolved constituents of the HCl extracts from mounds, Mg about 10 to 20 %, and K about 8 to 24 %. By contrast, the total dissolved constituents from soil without termites consist mainly in Fe from 5 to 62 %, Al from 6

Table 2: Chemical composition of the extracted amorphous phases

Elements[*]	Sample pair 1		Sample pair 2		Sample pair 3		Sample pair 4	
	TM	SWT	TM	SWT	TM	SWT	TM	SWT
Si	751	1222	136.4	5.38	14.1	7.52	20.6	8.49
Al	4460	4275	370.1	39.4	34.7	30.6	112.0	70.3
Fe	5467	5170	bdl	0.04	bdl	bdl	0.07	0.04
Mn	135	16.9	18.6	0.17	8.59	25.2	2.23	2.41
Total	10813	10684	525.0	45.0	57.4	63.4	134.9	81.3

TM = termite mounds, SWT = soils without termites, bdl = below detection limit;
[*] The concentrations are in μg/g

Table 3: Chemical composition of the <2 μm clay fractions

Elements[*]	Clays of termite mounds	Clays of soils without termites
Si	219.9	224.9
Al	104.0	103.1
Mg	3.10	2.04
Ca	3.90	0.52
Fe	63.1	72.2
Mn	0.46	0.22
Ti	9.48	9.61
Na	0.33	0.09
K	7.76	5.69
P	1.29	0.74
Rb	27.6	28.6
Sr	45.0	30.0
Sr/Ca	11 (10^{-3})	57 (10^{-3})
K/Rb	282	198
La	24.2	29.4
Ce	47.6	50.7
Pr	4.29	5.23
Nd	14.2	17.2
Sm	2.31	2.87
Eu	0.51	0.64
Gd	1.65	2.05
Tb	0.25	0.31
Dy	1.43	1.83
Ho	0.28	0.36
Er	0.8	1.01
Tm	0.14	0.17
Yb	0.86	1.09
Lu	0.14	0.17
Total	98.6	112.9

[*] The concentrations are in mg/g for the major elements and in μg/g for the trace elements and the lanthanides

to 61 %, and Ca from 5 to 38 %. The K/Rb ratio of the HCl extracts from mounds ranges from 13 to 320 and that from the soils without termites from 6 to 111. The HCl extracts also differ from each other in the Sr/Ca ratio of the two types of samples: those from termite mounds range from 6 to 151 x 10^{-3} and those from soils without termites from 8 to 1537 x 10^{-3}. As a consequence of the termite activity, the HCl leachates are enriched in all elements except Al, which is found to be depleted in the mounds of the pairs 3 and 4, Ti and Na being depleted in the mound of pair 1 and Ti in the mound of pair 3. The HCl extracts also differ clearly in the total lanthanide concentrations and in the magnitude of the Ce anomaly among the sample pairs. Those from termite mounds have total concentrations of 16 to 366 μg/g of soil. The distribution of the data from termite mounds normalized to the PAAS reference have patterns with apparent enrichments in the middle lanthanides accompanied by negative Ce anomalies from 0.004 to 0.31 for the mounds 1 and 2 and a positive Ce anomaly of 1.54 in the mound 3. A small but significant Eu anomaly of 1.22 is seen for the sample from mound 3, the other Eu anomalies being not significant due to analytical uncertainty (Fig. 3). The HCl extracts from soils without termites yield total concentrations of lanthanides of 0.69 to 687 μg/g (Table 5). The distribution patterns are also characterized by enrichments in the middle lanthanides with a negative Ce anomaly in the soils with termites of the sample pairs 1 and 2, a positive Ce anomaly of 1.22 and 3.61 in the same soils of the sample pairs 3 and 4, and a significant positive Eu anomaly of 1.25 in the same soil of the pair 3 (Fig. 3).

Termite activity apparently produced a consistent pattern of decreases in Si concentration and increases in Al, Ti, Ca, K, Rb, and Sr concentrations in the soil materials. This systematic Si mobility has already been reported in studies on the lateritization process widely described in tropical soils, leaving Fe- and Al-hydroxides behind (e.g., Millot, 1964), under a condition of high soil alkalinity. Timofeev (1987) suggested that this high alkalinity primarily makes P available to termites and other living beings. Breznak and Brune (1994), Brune *et al.* (1995) and Brune and Kuhl (1996) further suggested that high pHs help termites to digest soil bacteria and to solubilize the polyphenolic compounds.
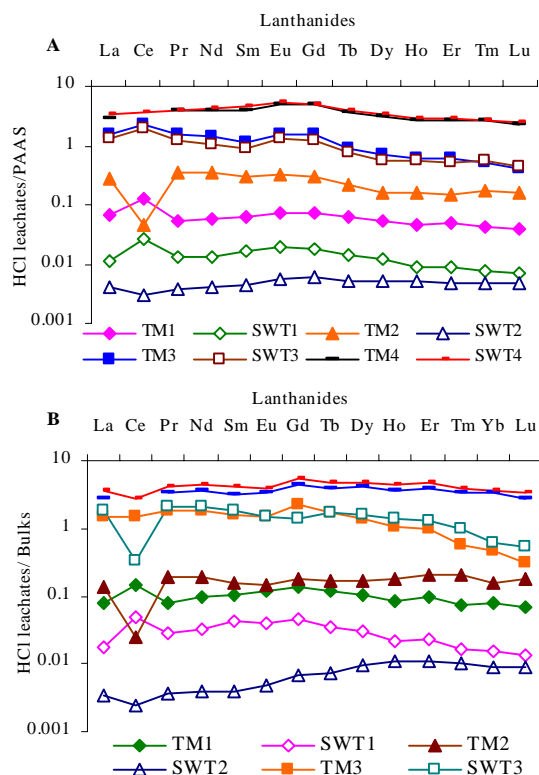
Fig. 3: Concentrations in lanthanides of the HCl leachates from termite mounds normalized (A) to the lanthanide pattern of the PAAS, and (B) to the lanthanide patterns in the corresponding mounds and soils called bulks. TM stands for termite mounds and SWT for soils without termites.

A potential cause for much of the elemental changes in soils hosting active termites is the relocation of many organic constituents from decomposed organic matter into framework minerals of the soils aided by the associated living system. Such relocations involve either nucleation and growth of new phases, or surface adsorption by termite modified minerals, or both. This is illustrated by the fact that the H_2O and HCl leachates of materials from termite mounds and corresponding soils without termites substantially differ from each other in their chemical characteristics. Termite activity reorganized Ca from soils to form part of the water and acid-soluble phases, possibly some poorly crystalline Ca-Mg carbonate phases and Ca-bearing phosphates.

Although part of the Mn increase observed in the HCl leachates may relate to precipitation of a carbonate phase, at least some of it may be linked to Fe oxy-hydroxides or Mn hydroxides. Birnessite or cryptomelane, for instance, are also known to

accommodate some K (Lu *et al.*, 2003; Rives *et al.*, 2004), which could account for the accompanying K increase that is observed in the HCl leachates. In fact, relocation of elements as a result of termite activity is most clearly evident in the changing contents of the trace elements and in the K/Rb ratio. While the mounds and corresponding soils differ little in their K/Rb ratios, the HCl leachates from the same samples are significantly different in their K/Rb ratios. It might be added that the K/Rb values obtained for the mounds and soils are typical for common soil-forming silicate minerals (Chaudhuri *et al.*, 2007). However, a relocation of K and Rb by an inorganic process alone, such as their reincorporation into newly-formed minerals after having been released during weathering of a primary mineral, does not explain the relatively high K/Rb ratio of about 1040 for the H_2O leachates from termite mounds. An intervention of a biochemical process is a potential explanation for the high K/Rb ratio. The solubility of silicate minerals enhanced by termite activity might have contributed to the production of Si-bearing amorphous phases with susceptibility to dissolution by H_2O and HCl, which may account for their higher concentrations in such solvent extracts. The concomitant significant increases of Al, Mg, Ca and P in termite mounds, as recorded in the H_2O and HCl leachates, are therefore responses to termite activity.

The K-Ar tracing

Among the <2 μm fractions of the three pairs of samples that were dated by the K-Ar method, that of one termite mound (pair 3) outlines a significant increase of 46.7 % in the K content, while the K content of the same clay size fraction of another mound (pair 1) is enriched by 70.8 %, and that of the third mound size fraction (pair 4) decreases by about 10 %. Termite activity leads to an increase in the radiogenic [40]Ar content for the mound 3, and decreases in the mounds 1 and 4. In all three cases that were investigated, termite activity leads to decreases in the K-Ar dates. Such decreases are generally explained by the effect of an alteration process, monitored mainly by the loss of radiogenic [40]Ar (e.g., Clauer and Chaudhuri, 1995). This explanation is not plainly applicable to this current study, as both loss and gain of radiogenic [40]Ar have been observed in the termite modified soil materials (Table 1). Basically, there must be a consistent reasoning for this, as it may be kept in mind that any K-Ar value, which results from ratio between the contents of K and

its radiogenic ^{40}Ar, integrates the individual apparent ages of all K-bearing soil particles. It may, therefore, be expected that soil clay particles are of diverse origin in terms of both, source and age. In other words, soil samples can certainly be regarded as heterogeneous material of varied provenance, with particles of different sizes and of varied types having different ^{40}Ar contents and K contents, which may explain the wide range of K-Ar apparent ages obtained on the clay fractions of either the termite mounds or the soils without termites. Groups of minerals characterized by different trace element compositions could potentially have different degrees of susceptibility to alteration or weathering.

Even very subtle, these differences in the trace element compositions may play a significant role in the alteration/weathering of the particles and, therefore, between mineral particles and organic materials. Some of the trace elements could, for instance, represent necessary cofactors for enzyme activities of microorganisms. The need for such elements would compel the microorganisms to seek the elements from particles. This would lead to selective destruction of minerals in a mass of heterogeneous clay particles, consequently leading to either an enrichment or a release of either radiogenic ^{40}Ar or K. The fact that gain of radiogenic ^{40}Ar in the clay fraction of one termite mound and loss in that of the two others draws a special attention towards the weathering process(es) involving clay minerals. In a broad view, weathering is seen as either a congruent or incongruent dissolution process.If the process is mainly congruent, the K-Ar apparent ages of the progressively altered/weathered K-bearing minerals should stay almost similar during the process as both isotopes are released concomitantly, which is not the case here, at least for two size fractions. In the case of a process dominated by an incongruent dissolution mode, the natural consequence is a decrease in the K-Ar data monitored by a preferential loss of radiogenic ^{40}Ar during the whole process (e.g., Clauer and Chaudhuri, 1995). Since this latter path is observed here in two out of the three studied sample pairs, it may be suggested that clay mineral alteration/weathering associated with termite activity is dominantly an incongruent dissolution process. Such a process of dissolution explains in turn, at least partly, the varied behaviour of the major and trace elements.

The variations of the K/Rb and Sr/Ca ratios

Whereas the bulk materials from termite mounds and from corresponding soils without termite activity differ little in their K/Rb ratios, the HCl leachates from termite inhabited materials clearly differs from HCl leachates from soils without termite activity, the former having systematically higher K/Rb ratios than the latter. The relative increase in the K/Rb ratio for the materials in the mounds is of such a magnitude that it can be explained in terms of selective adsorption/absorption of K over Rb by newly-formed solid products. But this explanation cannot rule out the possibility that the relatively higher K/Rb ratio for the mound materials arose at least in part as a consequence of the availability of these elements from plant materials that are being decomposed through termite activity. This may be suggested by the fact that water-soluble fractions from the materials of mounds have K/Rb ratios as high as 1040. Such a high ratio is common for plant materials and generally uncommon for common K-bearing silicate minerals (Chaudhuri *et al.,* 2007).

In summary, the main source for K and Rb of the dissolved phases in the HCl and H_2O leachates is not only the components of the clay-particle interlayer sites, even if the weathering of the mica-type components is incongruent as shown by the K-Ar data, but also the immobilized nutrients in the termites or in plants that termites bury in their mounds. However, as Rb is held more tightly than K in the interlayer sites of clay minerals, the K/Rb ratio of precipitated phases from a soil solution will be higher than the ratio of the same elements in the interlayer exchangeable position at equilibrium in K-bearing clay particles. The difference between K/Rb ratios of the HCl and H_2O leachates termite mounds and from soils without termites, could mainly be due to differences between the amounts of K and Rb released from both and it may reflect the modifications in the soil structure, as Jouquet *et al.* (2002) reported that termites can modify the mineralogical properties of some clays. Greatest differences in the Sr/Ca ratio were observed between the termite mounds and the soils without termites. Alternatively, the clay fractions and the corresponding termite mounds have nearly identical Sr/Ca ratios, these ratios being about 1/5 to 1/8 of the clay fractions and the corresponding soils without termites.

The termite activity apparently induced precipitation of H_2O and HCl extractable phase(s) having low Sr/Ca ratios. These phases could be amorphous or poorly crystalline Ca-phosphate or carbonate materials with low Sr contents, as evidenced by the extracts having middle lanthanide enrichments that are known to be associated with Ca-phosphate minerals. Since there is a correlation between the contents of Ca and P in the sample pairs 3 and 4, as well as in their H_2O and HCl leachates, the increase of P and Ca could be due to a precipitation of a phosphate phase related to termite activity. In the sample pairs 3 and 4, where the content of Ca is much higher in the termite mounds than in the soil without termites, the increase of Ca in the H_2O and HCl leachates may witness an important precipitation of carbonate phases compared to phosphate phases.

The behaviour of the lanthanides

Apparently, the termite activity induced an increase in the total concentration of lanthanides in all but one mound samples and in all but one HCl leachates. What should be noted here is that the lanthanide distribution patterns for the mound samples, relative to their corresponding soils without termite activity, are varied (Fig. 3). A concave upward distribution pattern, with middle lanthanide depletion, displayed by the mound 3 is an indication

that termite activity resulted in products with potential removal of phosphate-bearing phase(s), which are commonly known for their middle lanthanide enrichment. The presence of a Gd negative anomaly in relation to this particular mound sample (mound 3) is somewhat striking. Discount the possibility of any analytical error, in view of the fact that have observed Gd anomalies in several of studies which is involved plant material analyses. Therefore the Gd negative anomaly may in part reflect involvement of plant material lanthanides assimilated during chemical reorganization of soil minerals modified by termite activity. A second variation in the lanthanide distribution was recorded by both mound samples 2 and 4, each marked by a flat lanthanide distribution pattern with a negative Eu anomaly. This general lack of any fractionation among the lanthanides due to termite activity may imply that either the lanthanides were complexed by sulphate-bearing ligands or the lanthanides were mostly immobile rendering a very little loss of the lanthanide elements during modification of soil materials by termite activity. The Eu anomaly may suggest a variety of factors that might have come into play.

These include the influence of redox reaction since Eu is known to have both trivalent and divalent states, the influence of feldspar minerals which are known to have structural preference for divalent Eu

Table 4: Chemical composition of the H_2O extracts

Elements[*]	Sample pair 1		Sample pair 2		Sample pair 3		Sample pair 4	
	TM	SWT	TM	SWT	TM	SWT	TM	SWT
Si	74.7	17.6	346.0	8.47	14.3	15.0	26.7	5.59
Al	90.7	15.5	274.2	7.84	1.61	12.2	19.6	1.53
Mg	74.9	3.17	1320	4.69	4.68	1.31	0.44	0.47
Ca	164.9	2.93	3447	13.8	10.2	3.62	2.95	2.03
Fe	60.5	6.08	115.2	7.67	0.69	4.39	15.7	0.76
Mn	13.5	0.46	162.5	0.77	0.40	0.51	0.51	0.24
Ti	bdl	0.78	1.33	0.09	0.03	0.55	0.83	0.07
Na	bdl	0.41	483.0	3.43	2.11	1.15	2.26	0.74
K	266.4	9.71	8295	19.1	78.9	5.93	21.0	13.2
P	ND	ND	609.9	4.01	6.13	0.20	0.69	0.21
Total	746.0	57.0	15055	70	119.1	45.0	90.7	25.0
Rb	1452	60.3	28.0	0.16	0.08	0.04	0.09	0.10
Sr	1057	33.9	14.4	0.12	0.11	0.05	0.02	0.02
Sr/Ca (10^{-3})	6.4	11.5	4.18	8.60	11.1	13.5	6.6	10.3
K/Rb	184	161	297	120	1040	161	225	129

TM = termite mounds, swt = soils without termites, ND = Not Detected and bdl = below detection limit

[*]The concentrations are in μg/g

Table 5: Chemical composition of the HCl extracts

Elements*	Sample pair 1		Sample pair 2		Sample pair 3		Sample pair 4	
	TM	SWT	TM	SWT	TM	SWT	TM	SWT
Si	0.48	0.01	0.44	0.01	0.07	0.02	0.06	0.03
Al	6.75	0.39	0.28	0.07	0.02	0.02	0.06	0.07
Mg	25.5	0.02	15.8	0.01	0.26	0.04	0.09	0.02
Ca	75.7	0.03	90.0	0.06	0.63	0.14	0.29	0.07
Fe	7.36	0.15	2.63	0.63	0.06	0.02	0.18	0.11
Mn	7.59	0.01	2.85	0.01	0.15	0.10	0.08	0.03
Ti	bdl	bdl	0.01	bdl	bdl	bdl	bdl	bdl
Na	bdl	bdl	0.47	bdl	bdl	bdl	bdl	bdl
K	9.56	0.03	12.9	0.02	0.37	0.04	0.20	0.06
P	ND	ND	3.85	0.21	0.02	bdl	bdl	bdl
Total	133.0	0.64	129.2	1.02	1.58	0.38	0.96	0.38
Rb	29.9	0.26	44.03	0.38	6.39	3.41	15.6	9.99
Sr	797	0.38	562.12	0.47	94.5	25.7	34.7	110.4
Cr	6.34	0.25	bdl	0.71	bdl	bdl	bdl	bdl
Co	3.96	0.05	2.40	0.02	11.0	9.62	7.29	10.8
Ni	0.00	0.24	4.96	0.09	2.82	0.62	1.33	1.21
Cu	16.9	1.11	27.3	1.16	8.84	5.36	24.5	19.9
Zn	76.1	1.49	168.9	0.70	8.94	2.10	6.22	3.87
Rb	29.9	0.26	44.0	0.38	6.40	3.41	15.6	9.99
Sr	797	0.38	562	0.47	94.5	25.7	34.7	110.4
Zr			3.24	1.67	0.32	0.13	1.02	0.64
Mo	0.01	bdl	0.21	bdl	bdl	0.04	bdl	bdl
Cd	1.79	0.01	2.06	0.03	0.08	0.12	0.06	0.06
Cs			0.32	bdl	0.01	0.05	0.26	0.61
Ba	195	1.59	147.0	0.87	719	339	151.5	110.4
Pb	7.10	0.68	3.36	0.05	16.9	21.8	33.9	38.2
Th	1.26	0.09	0.04	0.02	0.13	0.13	0.94	0.57
U	0.38	0.09	0.16	0.12	0.61	0.61	5.78	5.13
Sr/Ca (10^{-3})	11	13	6	8	151	179	120	1537
K/Rb	320	111	293	61	58	10	13	6
U/Th	0.30	0.99	4.18	5.31	4.61	4.72	6.13	9.95
La	2.55	0.42	10.7	0.15	61.0	49.6	113.9	126.9
Ce	9.77	2.04	3.53	0.25	175.6	156.9	bdl	289.1
Pr	0.48	0.11	3.20	0.03	13.5	10.6	33.3	35.7
Nd	1.93	0.44	11.9	0.14	48.1	36.5	131.8	142.1
Sm	0.36	0.09	1.68	0.03	6.02	4.81	22.4	25.1
Eu	0.08	0.02	0.34	0.01	1.72	1.39	5.24	5.69
Gd	0.35	0.09	1.34	0.03	6.92	5.50	22.7	23.6
Tb	0.05	0.01	0.16	bdl	0.69	0.58	2.71	3.01
Dy	0.25	0.06	0.74	0.02	3.42	2.69	14.6	15.6
Ho	0.04	0.01	0.15	0.01	0.59	0.54	2.63	2.86
Er	0.14	0.03	0.43	0.01	1.74	1.51	7.56	8.31
Tm	0.02	0.00	0.07	bdl	0.21	0.22	1.05	1.10
Yb	0.12	0.02	0.34	0.01	1.26	1.08	6.86	7.06
Lu	0.02	0.00	0.07	bdl	0.17	0.19	0.97	1.08
Total	16.2	3.34	34.7	0.69	320.9	272.2	365.7	687.2
Ce/Ce*	0.06	0.01	0.31	0.004	1.54	1.22	ND	3.61
Eu/Eu*	1.08	1.11	1.08	1.06	1.24	1.25	1.09	1.10

TM = termite mounds, swt = soils without termites, bdl = below detection limit and ND = Not Detectable
* The concentrations are in mg/g for the major elements, in μg/g for the trace elements and the lanthanides

over trivalent Eu because of the close similarity in the ionic radius between the divalent Eu and Ca ions, and the influence of preferential transport of Eu involved in enzyme activated reactions. The influence of redox reaction may not have been very significant, as no correlation became evident between Fe depletion or Mn depletion and Eu depletion. The influence of feldspar may not be strongly argued

because the presence of any feldspar has not been directly observed, although the soils are known to contain a significant amount of kaolinite mineral which is often known to be a product of weathering of feldspar mineral. A third lanthanide distribution pattern is marked by light lanthanide enrichment. This may be explained in terms of preferential increased mobility of heavy lanthanides aided by high stability of these lanthanides building complexes with common organic ligands such as acetates, oxalates, malates etc., or with common inorganic ions such as bicarbonate and carbonate ions. The presence of Ce anomaly within the distribution patterns of lanthanides may be explained in terms of the presence of minor amounts of oxides and hydroxides of Mn and Fe, which are known to incorporate preferentially Ce over other neighbouring lanthanides.

The variations of the U and Th concentrations

As for most elements, the U and Th concentrations increased in the termite mounds. The increase for Th is relatively higher than that for U. In the HCl leachates, the decrease in the U and Th concentrations occurs in all mounds, except in that of the sample pair 3. As for the bulk samples, the U/Th ratios of the HCl leachates from termite mounds are lower than those of the HCl leachates from soils without termites. The increase in the U and Th concentrations in the mounds could be due to the incorporation of more U and Th in the newly-formed phases dissolved in HCl, relative to the soils without termites.

With the exception of the mound 2, the U/Th ratios of the soils without termites and the HCl leachates in the termite mounds were found to be higher than those of the soils and HCl leachates without termites. The decrease in these ratios because of a termite activity could be due to a fractionation effect during incorporation of U and Th in the newly-formed phases dissolved by HCl leaching. In presence of termites, some chelating agent could be more abundant than in the soil without termites, and make diffusion as a dominated mechanism for the incorporation of U and Th in the new phases in presence of termites.

CONCLUSION

This study showed that activity of termites induced significant chemical changes in the materials that they use to build their nests, increasing the contents of most major elements, as well as of U and Th, relative to the soils with no visible termite activity, except Si and to a lesser extent Fe, Mn, Na and K. A potential cause for such increases is the relocation of these elements into newly-formed H_2O and HCl leachable minerals. K-Ar results suggest an incongruent weathering of the clay material due to biological activity, explaining, at least partly, the varied behaviour of the major and trace elements. Ca and Mg dissolved in the two types of leachates whiteness for crystallization of Ca-Mg carbonate phases, part of the Mn probably relating to precipitation of a carbonate phase, and most to precipitation of Fe oxy-hydroxides and/or Mn hydroxides. Termite activity also induced precipitation of H_2O and HCl extractable phase(s) having low Sr/Ca ratios. These phases seem to be amorphous or poorly crystalline Ca-phosphates or carbonates, as evidenced by middle lanthanide enrichments that are known to be associated with Ca-phosphates. Even if the K/Rb values of the termite mounds are typical for common soil-forming silicate minerals, their relocation by an inorganic process alone does not explain an abnormally high ratio in the H_2O leachable mineral phases. It was also shown that the main source for K and Rb of the dissolved phases is not only the interlayer site of clay particles, but also nutrients immobilized in and by the termites. The termite mounds are especially enriched in light lanthanides relative to the corresponding soils without termites. The changing patterns support the precipitation of newly-formed mineral phases dissolved by dilute HCl and point to Mn-Fe oxy-hydroxides and Ca carbonates-phosphates. A mechanism by which increased amounts of Eu and Ce are linked to termite activity seems to relate to the occurrence of some reducing agent or agents that are released by the termites, causing reduction of Eu^{3+} to Eu^{2+} and Ce^{4+} to Ce^{3+} in a chelate, favouring in turn some selective incorporation of Eu^{2+} and Ce^{3+} in the new phases of termite modified soils.

The concentrations and patterns of lanthanides, and the K/Rb, U/Th and Sr/Ca ratios appear to be useful tools for studies of changes in soil chemical compositions. The studies of trace elements such as the lanthanides can also lead to increased understanding of dissolution/precipitation of phases produced by organic activity, and the consequent mobility of the released elements from

soils to ground- and surface waters. K-Ar isotopic dating appears also to be an appropriate tool to identify the dissolution processes of minerals.

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