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RESEARCH ARTICLE

CONTRIBUTION OF $\delta^{18}O$ AND $\delta^{13}C$ FROM SIDERITE TO THE HISTORY OF LAKE KAMALETE (CENTRAL GABON)

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Abstract

Located in the center of Gabon, in the South East of the Lopé National Park, Lake Kamalété has so far been the subject of several studies through the KAM1 core. We present here the study of the isotopic analyzes of $\delta^{18}O$ and $\delta^{13}C$ of the siderite crystals found in this core. This study provides new informations on the history of Lake Kamalété. Thus, the results indicate the existence of two dissolved inorganic carbon sources at the origin of the siderites present in the KAM1 core. This is atmospheric CO_2 for beige mud sediments (1410 to 520 years B.P) whose values of $\delta^{13}C$ are between -0.81 and +3.69 ‰, and CO_2 from methanogenesis for gray to gray green mud (from 520 years B.P to the present day) characterized by positive values of $\delta^{13}C$ between +3 and +9‰. These analyzes also indicate that Lake Kamalété has not known only a level of hydrological stability between 1255 and 520 years B.P but also an increase in evaporation during all its sedimentation marked by values of $\delta^{18}O$ ranging from -14.2 ‰ to -2.8‰.

Introduction:-

Stable isotopes of carbonates ($\delta^{18}O$ and $\delta^{13}C$) are a tool commonly used in paleolimnology through carbonate test organisms (foraminifera, ostracods) and carbonate minerals (calcite, aragonite, siderite) which record local paleoenvironmental variations (Talbot, 1990; Lu and Ku, 1997; Mayer and Schwark, 1999; Horton et al., 2015).

In Atlantic Central Africa, isotopic studies of carbonates are rare or even non-existent because the lacustrine sediments of this region are generally devoid of them. An exception is made for Lake Barombi-Mbo (Cameroon) whose sediments contain iron carbonates (Giresse et al. 1990) which have never been studied. Lake Kamalété located in central Gabon also revealed through the KAM1 core, the presence of diagenetic siderite crystals (Makaya M'voubou, 2005; Giresse et al, 2009) whose isotopic analyzes of $\delta^{18}O$ and $\delta^{13}C$ are studied in this article.The KAM1 core has already been the subject of several studies, in particular palynological (Ngomanda et al., 2007), sedimentological and biogeochemical (Makaya M'voubou, 2005; Giresse et al, 2009) then organic geochemistry (Sebag et al., 2013). These studies have highlighted two main paleoclimatic phases: a weakly humid phase ranging from 1350 to 520 years BP, then another wetter ranging from 520 years BP to the present. It should be noted that the weakly humid phase characterized by an opening of the landscape was disturbed by a short humid episode around 1350 years BP which resulted in the presence of pollen from forest species (Ngomanda et al., 2007).

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We propose in this paper to study for the first time the variations of the $\delta^{18}O$ and $\delta^{13}C$ of the siderite resulting from the sediments of a central African lake in this case, the Lake Kamalété. These isotopic data will be associated with the 14C datings already published by Giresse et al. (2009). This study is a contribution to the history of this lake.

Study setting:-

Lake Kamalété (0°42'S – 11°46'E) is located in central Gabon, at the South East of the Lopé National Park (Fig. 1). It lies in a narrow structural depression the francevillian rocks (Giresse et al., 2009). In the rainy season, the lake is 50 to 100 m wide, 500 to 700 m long and about 2 m deep. These values almost halve in the dry season. On the eastern and western sides of this lake, slopes of 20-40° are observed, which favor some slumping processes (Giresse et al. 2009). All of the flanks remain covered by a forest/savanna mosaic. This whole ecosystem is maintained by annual rainfall which is among the lowest in Gabon, ca. 1500 mm. This is because of rain shadow effect generated by the presence of the Massif du Chaillu and Mont Brazza which partially prevent the clouds laden with humidity from precipitating in the Lopé National Park.

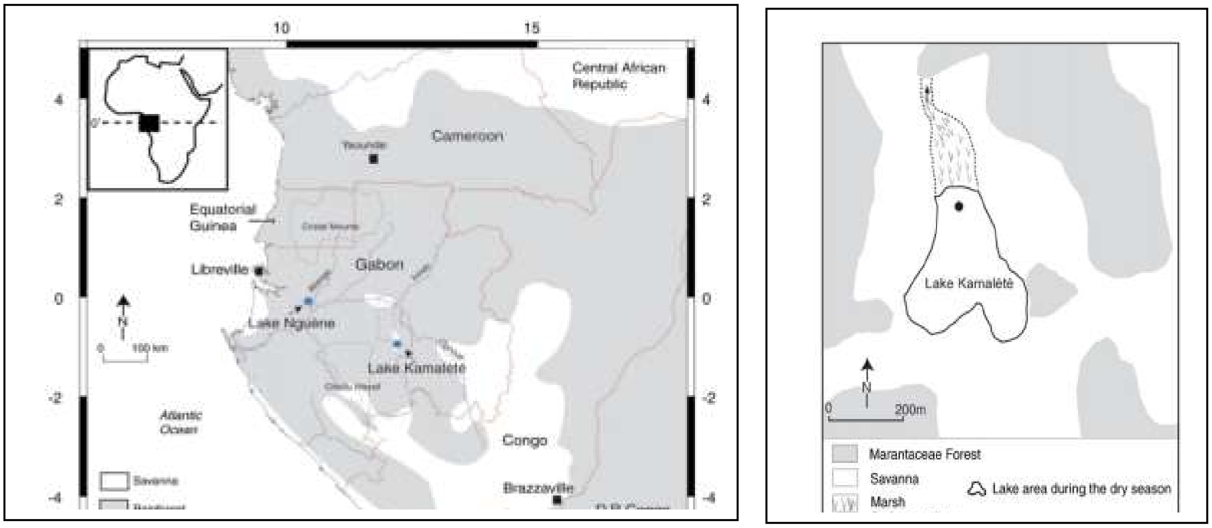

Fig. 1: Location of lake Kamalété (A) and core KAM 1 (B) (Ngomanda and al., 2007).

Material and Method:-

Sampling:-

The KAM1 core was taken from Lake Kamalété using a vibro-corer under a low water depth of approximately 1 meter. With a thickness of 371 cm, this core is made up of two distinct lithological units (Fig. 2), namely a beige vase ranging from 371 cm to 149 cm and a light gray vase ranging from 149 cm to the top of the core (Makaya M'voubou, 2005). Analyzed diagenetic siderite crystals (Photo) are continuously observed in the beige mud and then at certain depths in the light gray mud.

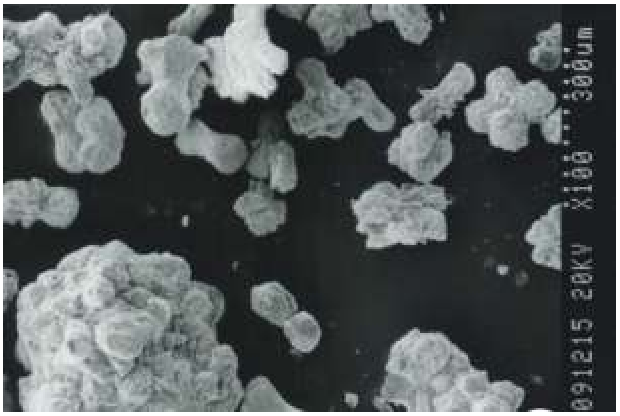

Photo : Siderite crystals in aggregates found in the core KAM 1.

Extraction of the siderite consisted of taking raw samples along the core using a global step of 10 cm. Then, each raw sample was washed through a 50µm sieve in order to obtain a pelitic fraction (<50µm) and a sandy fraction (>50m) in which siderite crystals were often presented with quartz grains and plant debris (Giresse et al. 2009). A total of 31 siderite samples were analyzed.

Treatment protocol:-
The analysis method consisted of measuring the stable oxygen ($\delta^{18}O$) and carbon ($\delta^{13}C$) isotopes of siderite crystals from the KAM1 core. These crystals were ground and the powder obtained was placed in a tube containing 10% hydrogen peroxide in order to eliminate organic matter likely to bias the measurements. After stirring the mixture for 2 hours, it is centrifuged to separate the powder from the hydrogen peroxide which is emptied. The powder residue remaining in the tube is rinsed with distilled water then dried in an oven at 90°. After drying, a few tens of micrograms of siderite powder are introduced into the mass spectrometer. The analysis of the powder is carried out by comparison with the N.B.S 19 (National Bureau of standard 19) which is an international standard issued by the Atomic Energy Agency. Its values are fixed with the Pee Dee Belemnite (P.D.B). The comparison thus makes it possible to calculate the isotopic compositions of $\delta^{18}O$ and $\delta^{13}C$ of the siderite expressed in per thousand compared to the N.B.S according to the following formulas:

$$\delta^{13}C = \left(\frac{R_{sample}}{R_{N.B.S}} - 1 \right) x1000 \text{ and } \delta^{18}O = \left(\frac{R_{sample}}{R_{N.B.S}} - 1 \right) x1000$$

$$R_{sample} = \left(\frac{^{13}C}{^{12}C} \right)_{sample} ; R_{N.B.S} = \left(\frac{^{13}C}{^{12}C} \right)_{N.B.S} ; R_{sample} = \left(\frac{^{18}O}{^{16}O} \right)_{sample} \text{ and } R_{N.B.S} = \left(\frac{^{18}O}{^{16}O} \right)_{N.B.S}$$

These analyzes were carried out at the University of Montpellier II.

Resultats:-

Figure 2 presents the results of isotopic analyzes of siderite ($\delta^{18}O$ and $\delta^{13}C$) associated with ^{14}C dating. These results indicate that:

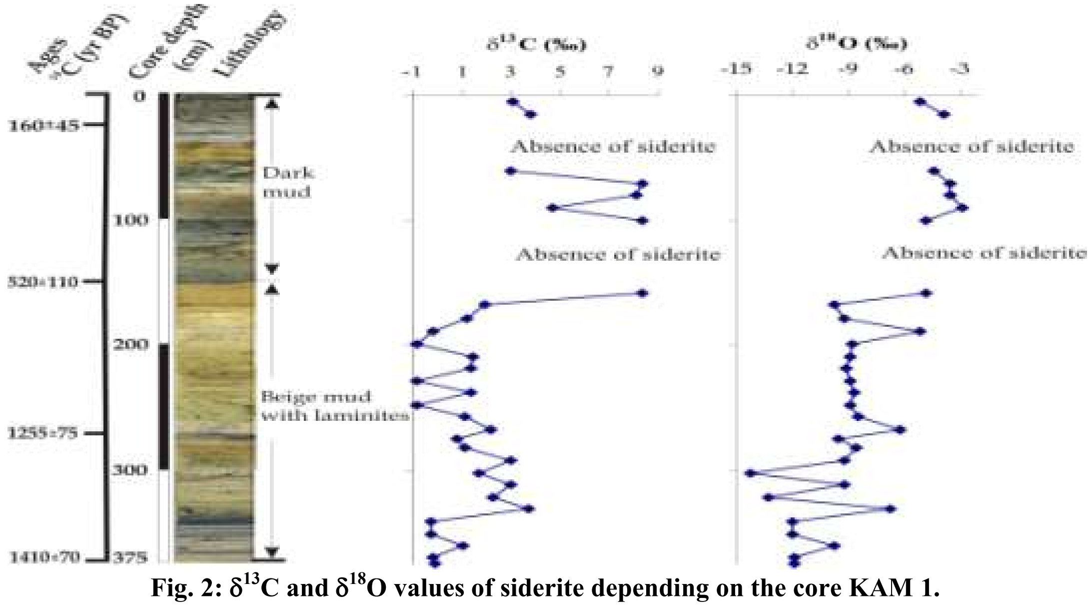

Fig. 2: $\delta^{13}C$ and $\delta^{18}O$ values of siderite depending on the core KAM 1.

- the values of $\delta^{13}C$ for siderite vary between -0.8 and +3.69 ‰. In the beige mud (1410 to 520 years B.P), the $\delta^{13}C$ values of the siderite fluctuate enormously between -0.81 and +3.69 ‰ whereas they aare all positive (between +3 and +9‰) in the light gray mud ranging from 520 years B.P to the present day.
- the $\delta^{18}O$ values of the siderite are negative over the entire core and between -14.2‰ and -2.8‰. In the beige mud, the $\delta^{18}O$ values of the siderite fluctuate enormously between -14.2 and -6.22 ‰ starting from the base of the core down to 2.68 m. Then, they stabilize around -9 ‰ between 2.58 and 1.68 m despite a peak recorded at 1.9 m

corresponding to the value of -5 ‰. In the light gray mud, the $\delta^{18}O$ values of the siderite are higher than -4.8 ‰ and therefore higher than those of the first of the deposit. In general, these values show an increasing trend from the base to the top of the deposit.

Figure 3 indicates some weak correlations between $\delta^{13}C$ and $\delta^{18}O$ of the siderite on the beige mud (3.3 to 1.68 m) ($R^2 = 0.08$) and on the whole dark mud ($R^2 = 0.06$) but this one is very strong ($R^2 = 0.98$) between the base of the core and 3.3 m of depth.

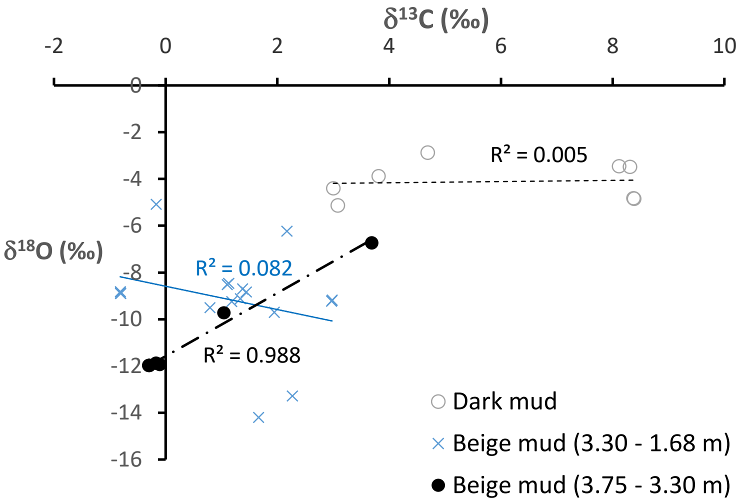

Fig. 3: $\delta^{13}C$ and $\delta^{18}O$ of siderite correlation depending on the lithologic levels of the core KAM 1.

Discussion:-

The interpretation of variations of $\delta^{13}C$ and $\delta^{18}O$ of lacustrine carbonates is rarely easy (Uysal et al., 2000) because these are controlled by several paleoclimatic factors whose significance can be local, regional or global. The factors in question are specific to each of the isotopes and have been described by several authors (Talbot, 1990; Li and Ku, 1997; Mayer and Schwark, 1999).The results of the isotopic analyzes of $\delta^{13}C$ and $\delta^{18}O$ of siderite crystals from Lake Kamalété were presented according to the two lithological sets, namely the beige mud ranging from 1410 to 520 years B.P and the gray to gray green mud ranging from 520 years. B.P to the present day.In the beige mud (1410 to 520 years B.P), the values of $\delta^{13}C$ are between -0.81 and +3.69 ‰ These results reveal that atmospheric CO_2 is the source of dissolved inorganic carbon having prevailed during the precipitation of siderite crystals (Mayer and Schwark, 1999). It can therefore be said that siderite was formed in equilibrium with atmospheric CO_2. This must have been made possible thanks to a low lake level which facilitated exchanges between atmospheric CO_2 and sediments. According to Makaya M'voubou (2005), the period from 1410 to 520 years B.P is generally characterized by a slice of water much thinner than that which the lake recorded between 520 years B.P and the present day.

In the gray to gray-green mud, the values of $\delta^{13}C$ between +3 and +9‰ are more positive than the previous ones. They are associated with a change of facies. Indeed, the gray to gray-green facies of this vase corresponds to a more anoxic environment and therefore more reduced than the previous one. This would explain the discontinuous formation of siderite crystals in this part of the deposit. Also, the values of $\delta^{13}C$ fluctuating between +3 and +9‰ indicate the appearance of a new source of dissolved inorganic carbon at the origin of the formation of siderite crystals within this mud. This source would probably be linked to methanogenesis. Indeed, during the opening of the core, we observed signs of CO_2 release in this mud linked to this phenomenon. According to Bahrig (1989), the precipitation of siderite crystals during methanogenesis is characterized by high $\delta^{13}C$ values which can reach +10‰. This change in the source of dissolved inorganic carbon is correlated with the rise in the level of the lake, which corresponds to the establishment of the gray to gray green facies (Makaya M'voubou, 2005; Giresse et al., 2009).
Concerning the $\delta^{18}O$ of the siderite, these values are negative and its evolution showed an increasing trend with a difference of 6 ‰ between the base and the top of the core.

The $\delta^{18}O$ curve makes it possible to distinguish three episodes linked to the hydrological variations of the lake. The first and second episodes correspond to the beige mud which attests to a low lake level resulting from a low humidity climate (Makaya M'voubou, 2005; Giresse et al., 2009). On the other hand, the third episode corresponds to the gray to gray-green mud which testifies to a rise in lake level following a humidification of the climate according to the same authors.The first episode (1410-1255 years BP) shows regular variations of $\delta^{18}O$ of siderite which can be likened to small fluctuations in the level of Lake Kamalété while it remains low due to the scanty rainfall (Giresse et al., 2009). The strong correlation (R^2=0.98) between $\delta^{13}C$ and $\delta^{18}O$ highlighted between the base of the core and 330 cm depth indicates according to Li and Ku (1997) that lake was closed before 1255 years BP. At that time, the current outlet was therefore not working.

The second episode (1255-520 years BP) is marked compared to the previous episode by a relative increase then, an overall stability of the values of $\delta^{18}O$ of siderite around -9‰. This could correspond, despite the scanty rains, to a slight increase in lake level and then to its stability. During this period, the absence of correlation (R^2=0.08) between $\delta^{13}C$ and $\delta^{18}O$ attests to the opening of the lake and therefore the functioning of the lake outlet.Finally, the third episode (from 520 years BP to the present day) corresponds to the gray to gray-green mud. It is characterized by siderite values of $\delta^{18}O$ which vary very little and are higher than those of the previous episode. These values are associated with a rise in lake level resulting from the humidification of the climate (Makaya M'voubou, 2005; Giresse et al., 2009). Here too, there is an absence of correlation between $\delta^{13}C$ and $\delta^{18}O$ (R^2 = 0.006) which is consistent with the current operation of the outlet.The trend of increasing siderite $\delta^{18}O$ values observed from the base to the top of the core reflects an increase in evaporation during deposition. This is due to the insolation which is stronger in the humid phase (third episode) and less in the weakly humid phase (first and second episodes). Indeed, during the strong insolation, the ^{12}C (lighter) of the inorganic carbon dissolved in the water of the lake, evaporates in the atmosphere to the detriment of the heavier ^{13}C which remains in abundance in lake.

Conclusion:-

Analyses of $\delta^{13}C$ and $\delta^{18}O$ carried out on siderite crystals from Lake Kamalété provide new information on the history of Lake Kamalété. They indicate that:

- Atmospheric CO_2 is the main source of dissolved inorganic carbon at the origin of siderite precipitation in the beige mud (from 1410 to 520 years B.P). A new source appeared during the formation of the siderites of the gray to gray green mud (from 520 years B.P to the present day): it is the CO_2 of methanogenesis;
- Lake Kamalété experienced a level of hydrological stability between 1255 and 520 years B.P;
- Evaporation has increased over time.

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