

## $\delta^{18}\text{O}$ and $\delta\text{D}$ variations in some volcanic lakes on the Cameroon Volcanic Line (West-Africa): generating isotopic baseline data for volcano monitoring and surveillance in Cameroon

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### ABSTRACT

Based on geo-anthropological and geochemical studies, catastrophes similar to the unprecedented gas explosions in the mid-1980s from the Cameroonian killer lakes Nyos and Monoun, might occur in any of the 37 other lakes located along the Cameroon Volcanic Line (CVL). Because people could suffer loss and desolation from predictable catastrophes in the future, monitoring/surveillance policies must be established. Due to their location, crater lakes integrate the geochemical processes that develop in the Earth's crust due to magmatic activities. Therefore, monitoring the surface manifestations of those deep seated and/or hydrothermal processes might reveal increases/decreases in magmatic activities. The anomalous changes in a volcanic lake induced by mixing with exogenous fluids that have a specific  $\delta^{18}\text{O}$  and  $\delta\text{D}$  compositional fingerprint (magmatic, metamorphic, etc.) could be utilized to predict volcanic hazards. If the steady state of a lake environment and the external and intrinsic parameters that control its hydrodynamics are clearly identified and reasonably understood, the anomalous evolutionary processes that compromise its stability can be identified. This study attempts to collect the  $\delta^{18}\text{O}$  and  $\delta\text{D}$  data from 17 Cameroonian lakes to help establish a volcano-related monitoring/surveillance network. This work identifies the processes that control the isotopic composition of the lakes and assesses the intra-/inter- and spatial  $\delta^{18}\text{O}/\delta\text{D}$  variations. Almost all of the lakes contain meteoric water. These lakes are mostly isotopically stratified; epilimnia is generally more positive than the hypolimnia. However, although the rainfall is gradually depleted in heavy isotopes when moving from the South to the North due to the latitude effect, the lakes become more enriched (0.6‰/100 km) due to evaporation. The evaluated impact of several parameters on the isotopic variation suggests that the hydrological setting may play an important, albeit not preeminent, role relative to the other factors. Consequently, the interplay between climatology, phytogeography, hydrology and morphometry might help shape the isotopic composition of the lakes.

Key words: Epilimnia, hazard, Cameroon, Lake Nyos, Lake Monoun, gas explosion.

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### INTRODUCTION

Due to their peculiar setting, crater lakes attract attention worldwide. In addition to their aesthetic and recreational benefits (Butler, 1991; Eagles, 1997), these lakes play important economic roles such as acting as a water source for domestic use, hydroelectric power production and reservoirs for irrigation. Their global importance is also obvious: they house a remarkable variety of living organisms, making them a reservoir of biodiversity; they also participate in the global carbon cycle because they can passively and/or explosively release a large amount of  $\text{CO}_2$  into the atmosphere (Allard *et al.*, 1991; Freeth, 1992; Perez *et al.*, 2011), thus participating in global climate regulation.

However, aside from these advantages, the presence of a volcanic lake might negatively impact the neighboring populations and surrounding areas. For example, these lakes can pollute their immediate environment, as observed during the acidic contamination of Ciwidey River network in Indonesia, which damaged agricultural soils (Sriwana *et al.*, 1998; van Rotterdam-Los *et al.*, 2008; Rouwet *et al.*, 2014). These lakes are considered hazardous due to the recurrent and devastating lahars, avalanches (O'Shea, 1954; Blong, 1984; Badrudin, 1994) and phreatic eruptions (Rowe *et al.*, 1992; Christenson, 2000; Christenson *et al.*, 2010) associated with them. In addition, lakes Nyos and Monoun revealed a new type of volcanic lake-related hazard. In the mid-1980s, the two lakes instantaneously re-

leased copious amount of CO<sub>2</sub> to the atmosphere, killing approximately 1800 people (Kling *et al.*, 1987; Sigurdsson *et al.*, 1987; Tazieff *et al.*, 1987; Kusakabe *et al.*, 1989; Evans *et al.*, 1994; Kling *et al.*, 2005). The *killer lakes* tragedies spurred researchers to survey crater lakes in connection with this new geologic hazard in locations, such as Italy (Martini *et al.*, 1994; Cabassi *et al.*, 2013), Indonesia (Christenson, 1994) and Costa Rica (CVL7 Workshop). Based on recent geochemical studies, Nyos- and Monoun-type events are recurrent phenomena (Kling, 1987b; Tietze, 1992; Halbwachs *et al.*, 1993, 2004; Schmid *et al.*, 2004; Kling *et al.*, 2005; Kusakabe *et al.*, 2008) and may not be as unique as the extraordinary events associated with volcanic lakes that might have occurred elsewhere in the Cameroon Volcanic Line (CVL) corridor, according to social studies (Shanklin, 1992); in particular, the lake-related *maleficent* activities that brought death and destruction to some ethnic groups, such as the Bamessi, Kom and Oku in the Oku region, forced these populations to emigrate (Shanklin, 1992; Eby and Evans, 2006). Although the CVL lakes are obviously serious threats, they have rarely been studied (Hassert, 1912; Corbet *et al.*, 1973; Green *et al.*, 1973; Kling, 1987a; Tanyileke *et al.*, 1996) to evaluate the risks for the neighboring populations.

Based on recent studies, Lake Monoun may release CO<sub>2</sub> approximately every 30-40 years, while Lake Nyos requires a longer interval, which is approximately 100 years (Kusakabe *et al.*, 2008; Issa *et al.*, 2013). Despite the historical and geochemical evidence, few or no records describing similar events among the populations living in the Nyos and Monoun areas have been discovered (Eby and Evans, 2006). One hundred years is a relatively a short period on the societal time scale for a given community to forget about such unique and deadly events because the 17<sup>th</sup> century volcanic eruption of Long Island off the coast of Papua New Guinea was well preserved through oral transmission (Blong, 1984). Therefore, the lack of clues supporting a history of catastrophic events in the areas around lakes Nyos and Monoun may imply the following: i) gas accumulation is a relatively recent phenomenon; ii) the limnic eruptions in the 1980s might have occurred when tectonic events created and/or reopened sublacustrine vents/faults, allowing gas to seep into the lakes; iii) the gas input rate has dramatically increased due to a similar increase in magma degassing in recent years. In summary, the geochemical and social studies described above suggest that any of the approximately 37 CVL lakes (Kling, 1987a), especially those able to accumulate a harmful amount of gas (Freeth, 1992), could host Lake Nyos- and Lake Monoun-type events or other crater lake-related hazards. Limnic eruptions are unique, and the circumstances required for these events are rare (Kling *et al.*, 1987). The debate remains open. However, the geochemical and historical evidence pre-

sented above suggest that future catastrophes along the CVL cannot be completely ruled out. Therefore, establishing a minimal monitoring/surveillance network for the CVL lakes is the only way to prevent death and sorrow.

To prevent volcano-related hazards in general and crater lake-related hazards in particular, regularly monitoring the physicochemical properties of these structures might reveal anomalies that predict impending catastrophes through temporal variations. After the report by Kling (1987a), only Lakes Nyos and Monoun have been monitored continuously. Little attention has been paid to the other crater lakes. Studying these lesser-known lakes will provide an assessment of their present status while generating the geochemical data needed to assess their future evolution; these data will help predict any dangerous developments, enabling the application of the appropriate mitigation measures because, some (*e.g.*, Lakes Tizon, Barombi Mbo, Wum, Oku, Manenguba twin lakes, Enep) are located in densely populated areas. In addition, after the Lake Nyos and Lake Monoun catastrophes, the neighboring populations have lived in a state of psychological unrest. This state of unrest is obvious based on the frequent reports of *unusual* events, such as the overturn of Lake Barombi Mbo in the 1940s and 2012 and Lake Wum in 2001, or extreme meteorological events such as the water spout in Lake Oku in 2011 (Issa *et al.*, 2011; *Institute for Geological and Mining Research (IRGM) internal report, unpublished*).

This study discusses the isotopic composition of several lakes located on the CVL. These lakes are distributed along an approximately 650 km WSW-ENE trending transect from 3°47'N at the edge of the Atlantic Ocean to 7°26'N in the Adamaua Plateau (Fig. 1). They range in elevation from approximately 40 m to approximately 2267 m asl (Tab. 1) and are grouped into three districts (Kling, 1987a): the Adamau Lake District (Ad-LD), the Bamenda Lake District (Bd-LD) and the South-west Lake District (SW-LD) (Fig. 1). Previous work using the stable isotopes of water encompassed five lakes (Kling *et al.*, 1987; Kusakabe *et al.*, 1989; Tanyileke *et al.*, 1996; Nagao *et al.*, 2010). Only the last two contained isotopic profiles for Lakes Nyos, Monoun and Wum without discussing their variations and controls further. The new data will assess reservoir dynamics (Brown *et al.*, 1989; Ohba *et al.*, 1994). These insights will enable us to understand and interpret any changes that may occur along the water column in response to an inflow of fluids with a distinct δ<sup>18</sup>O/δD composition. Those signals could reasonably be associated with the origin of these fluids, enabling investigations of the biogeochemical processes involved through tracers, such as the stable isotopes of water (IAEA, 1981; 1991). For example, the anomalies created by sinking cold rainwater might induce the onset of the gas blasts. In addition, detecting and assessing the mixing process of the lake with connate, formation, hydrothermal or magmatic fluids may

signal an increase in the sub-surface activities (Rowe *et al.*, 1992; Giggenbach, 1997; Rouwet *et al.*, 2014). Accordingly, this study aims to use (together with physico-chemical data) water isotopes as hazard indicator for the CVL lakes by providing a comprehensive overview of their spatial and intrinsic variations; this work generates isotopic data to complement other geochemical tools for volcano-related hazards prediction in Cameroon. Specifically, this study investigates the intra/inter-lake and spatial isotopic variations across 17 lakes (Tab. 1) and identifies the parameters that control those variations.

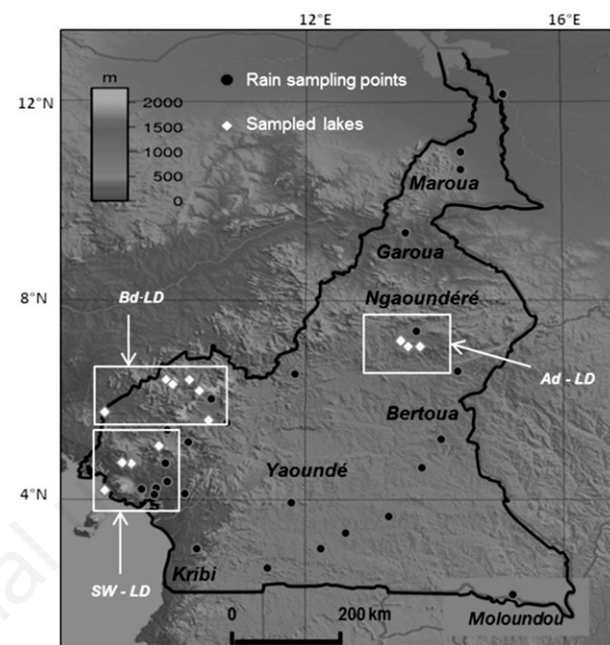
This work is one of the studies initiated within the framework of the Science and Technological Research Partnership for Sustainable Development (SATREPS) project entitled *Magmatic Fluids input in lakes Nyos and Monoun and Mitigation of Volcanic Hazards through Capacity Building in Cameroon* funded by the Governments of Japan and Cameroon. The SATREPS-NyMo (SATREPS Nyos-Monoun) intends to realize better safety for the Nyos and Monoun populations to rehabilitate the two areas affected by the mid-1980 gas explosions both economically and socially. Beyond the target areas, the data should reduce the risks posed by volcanic lakes in particular and volcanogenic structures in general.

## METHODS

### Geology, climatology and phytogeography

Cameroon is located on the coast of West Africa between 8° and 16° longitude East and 1.7° and 13° latitude North (Fig. 1). The CVL is a 1600 km long chain of ex-

tinct Cenozoic volcanoes, except the Mount Cameroun, which erupted three times within the last 30 years (Tsafack *et al.*, 2009). The geological makeup of the CVL



**Fig. 1.** The study area: digital elevation map (DEM) of Cameroon showing locations of rain samples (black circles) and location of the studied lakes (white rhombi). The three Lakes District (LDs) are also shown.

**Tab. 1.** Geographic positions and some characteristics of the studied lakes.

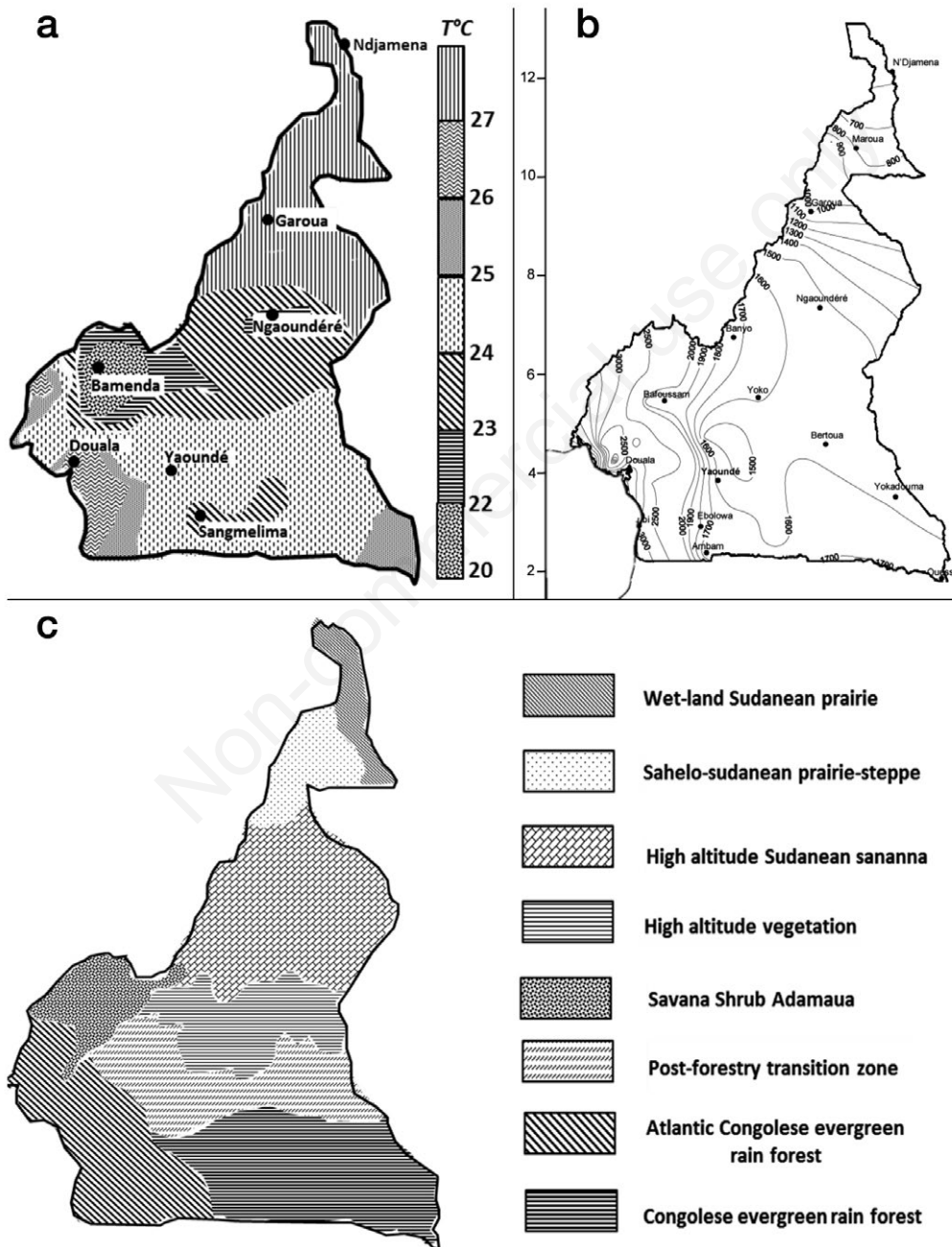
Lakes District	Name	Lat (°)	Long (°)	Elevation (m asl)	Area (ha)	Depth (m)	D (km)	avg. T (°C)	P (mm)	Type	Abb.
SW-LD	Barombi Koto	4.28	9.16	106	140	5.5	42.1	24.5		T-f	BK
	Barombi Mbo	4.65	9.41	301	415	110	65.5	24.5		T-f	BM
	Debunsha	4.06	8.98	54	6	13.5	0.7	26.5	3500	C	Db
	Manenguba Female	5.04	9.82	1920	22	168	127	23.5		H/T	Mf
	Manenguba Male	5.04	9.82	1900	2	92	127	23.5		H/T	Mm
Bd-LD	Baleng	5.33	10.26	1374	8	52	235	23.5		C	Ba
	Benakuma	6.26	9.57	576	154	138	288	23.5		T-f	Be
	Idjagham	5.75	8.99	100	70	17	165	25.5		C	Id
	Elum	6.20	10.02	960	50	35	251	22.5	2000	C	El
	Nyos	6.27	10.18	1091	158	208	278	22.5		T-f	Ny
	Monoun	5.35	10.35	1080	53	96	241	23.5		T-f	Mo
	Wum	6.41	10.06	1177	45	124	266	22.5		T/H	Wu
	Oku	6.12	10.27	2227	243	52	259	19		C	Ok
Ad-LD	Baledjam Marbuwi	7.08	13.52	1180	20	104	614	21		C	B-m
	Ngaoundaba	7.13	13.69	1160	10	62	620	21	1500	H/T	Ng
	Baledjam (Ranch)	7.08	13.52	1249	25	13	636	21		C	B-r
	Tizon	7.25	13.58	1160	8	48	612	21		C	Ti

*D*, distance from the Atlantic Ocean; *avg. T*, averaged temperature (estimated from Fig. 1b); *P*, precipitation (estimated from Fig. 1c); *Type*, hydrological setting; *SW-LD*, South-west Lake District; *Bd-LD*, Bamenda Lake District; *Ad-LD*, Adamau Lake District; *C*, closed lakes; *T-f*, through-flow lakes; *H/T*, head or terminal lakes; *depths and areas* from Kling, 1987a.

comprises sedimentary, metamorphic, plutonic and several volcanic series (Laclavière and Loung, 1979; Fitton and Dunlop, 1985). The basement rock complex is a mixture of gneiss, granites and micaschists with an age of 2500 to 1800 Ma (Fitton and Dunlop, 1985; Ngako *et al.*, 2008). All the CVL lakes, except Lake Idjagham, an as-trobleme formed in sedimentary terrain (Kling, 1987a),

are hosted in volcanoes formed in trachytic and/or recent basaltic series overlain by pyroclastic materials.

Because of its latitudinal extension, Cameroon is very diverse (Fig. 2). Fig. 2 a, b and c summarize the meteorological and phytogeographical characteristics across the territory which can be subdivided in three climatological domains: the Atlantic coast, with a hot and humid climate,



**Fig. 2.** Climatology and phytogeography. a) Temperature distribution across the territory. b) Overview of rainfall across the country. c) Phytogeography description. (Modified from Sighomnou, 2004).

has a mean annual temperature and rainfall of 26°C and 3000-4000 mm, respectively (Fig. 2 a,b). The relative humidity is about 89% throughout the year. On the Atlantic side of Mount Cameroon, the annual rainfall reaches *ca.* 12,000 mm (Fontes and Olivry, 1976) (Fig. 2b). The vegetation cover is characterized by evergreen forest and mangroves (Letouzey, 1968, 1985) (Fig. 2c). To the North, in the Adamau Plateau (mean elevation is *ca* 1500 m *asl.*), the mean temperature is 21°C, and the mean annual rainfall is *ca* 1500 mm (Fig. 2 a,b). The vegetation cover is of the sudano-Guinean type (White, 1983) and constitutes a transition between the green forest in the South and the savanna-steppe in the North (Fig. 2c). Between the coastal region and the Adamaua Plateau, lays the mountainous grassland with an altitude ranging from 700 to 2400 m *asl.* The mean temperature in the region is about 20°C (Fig. 2a). The rainy season delivers about 2000 mm  $y^{-1}$ . The evergreen highland type forest characterizes the vegetation in that region (Fig. 2c). However, the vegetal cover has been significantly altered by human activities in that highly populated area (Letouzey 1968, 1985; Suchel, 1987). Alike the climatology, the phytogeography and the geology, the hydrological setting of the lakes is also varied. Generally, lake water derives from precipitation on their surface, stream flow or in some setting, groundwater inflow. They lose water by evaporation, stream outflow and underground seepage. The mass balance between the inflows/outflows often results in change in storage, which in the CVL lakes, is reflected by a fluctuation in lake level of *ca.* 1-2 m  $y^{-1}$  as evidence by the visual landmark line on almost all the crater walls.

Based on the hydrological setting, the lakes can be grouped into three (Peterson *et al.*, 1999): The first group comprises the topographically isolated and often enclosed lakes characterized by no or very limited topographic basin tributaries; they gain water mostly from direct rainfall. However, because of the permeable nature of the hosting rocks and the volcanic deposit surrounding them, some groundwater may be transferred across the topographic divides. Of the 17 studied, lakes Tizon, Baledjam Marbuwi, Baledjam (ranch), Oku, Elum, Debunsha, Idjagham, Baleng, more or less share the above characteristic. A second group, the through-flow lakes, comprises lakes Nyos, Monoun, Barombi Mbo, Benakuma, and Barombi Kotto. They are hosted in river channels, thus, receive and lose water through rivers and/or seepage located at the contacts between the basement rock and the lava flows or through the poorly consolidated volcanic deposits. A third group composed of terminal and head lakes. The first ones are the ending point of one or several emissaries (Ngaoundaba, Manenguba Male and Female); the second, located upstream, constitute the rising point of perennial or ephemeral streams (*e.g.*, Lake Wum).

## Sampling and analytical techniques

Two sampling campaigns were conducted during the dry seasons of 2012 and 2013 (from March to May). The precipitation was sampled in Yaoundé, Bertoua, Ngaoundéré and Garoua. In addition, 18 occasional rainfalls were collected from the 12 sites during the 2012 and 2013 rainy seasons (Fig. 1). Tab. 2 lists the samples and their isotopic compositions. The lakes were investigated from the edge of the Atlantic Ocean (*e.g.*, Lake Debunsha) in the SW-LD to the Adamaua plateau in the Ad-LD (Fig. 1). The water samples were collected from the surface, middle depths and bottom using a 1.5 L Niskin bottle. Twenty-nine water samples were also collected from shallow wells, boreholes, soda springs, springs and outlet/inlet streams (Tab. 3). After sampling, the water was filtered through 45  $\mu$ m hydrophilic acetate syringe-driven filters in newly purchased 50 mL high-density polyethylene bottles that were rinsed three times with the water to be sampled before collection. The bottles were filled to the top and firmly capped to avoid evaporation. The sampled waters were analysed within one month using a Cavity Ringdown Spectrometer L-2120-i (Picarro Inc., Santa Clara, CA, USA) in the Laboratory of Volcanology and Geochemistry of Tokai University. The analytical precision was  $\pm 0.4\text{‰}$  and  $< \pm 0.1\text{‰}$  for  $\delta D$  and  $\delta^{18}O$ , respectively, and was expressed against VPDB.

The studied lakes were selected as follows: six lakes with maximum depths above 100 m, five with a maximum depth above 50 m and six with a depth  $< 50$  m were selected (Tab. 1). The first six lakes share the morphometric characteristics of lakes Nyos and Monoun. Therefore, they should also share some of their functional features, such as the stratification (strong) that prevents any mixing between the layers and the likelihood to store dangerous amount of gas; these studies may provide some insight into the material transport processes across the water column. The second group may also feature the same characteristics to a lesser extent. Finally, the last group includes lakes with little to no gas storage capacity, removing them as a serious threat. Studying these lakes shall provide insight into the isotopic characteristics of the remaining lakes.

## RESULTS AND DISCUSSION

### The Cameroon meteoric water line

Several studies of the water isotopes in Cameroon have been carried out using the global meteoric water line (GMWL) (Craig, 1961). However, since the stable isotopes in the precipitation of a given region may reflect specific meteorological and physical conditions (*e.g.*, Gat, 1996), they are better described by a local derivative: the local meteoric water line (LMWL). In a recent study, Garcin *et al.* (2012) proposed a Cameroon meteoric water

Tab. 2. Isotope ratios in precipitations with name of locations and source.

Site	Date	$\delta D$	$\delta^{18}O$	Ref.	Site	Date	$\delta D$	$\delta^{18}O$	Ref.	Site	Date	$\delta D$	$\delta^{18}O$	Ref.
Fontem	May 2012	1.06	-1.64	1	Yaoundé		27.2	1.30	6	Garoua		-49.4	-7.31	4
Monoun	Apr 2012	12.9	-0.62	1	Yaoundé		1.80	-1.87	6	Garoua		-70.4	-10.2	4
Nyos	May 2012	10.9	-1.12	1	Yaoundé		10.6	-0.97	6	Garoua		-66.3	-6.43	4
Buea	5.82	-0.72	2	Yaoundé		-13.4	-3.39	6	Garoua		-58.9	-8.98	4	
Buea	6.19	-0.81	2	Yaoundé		2.30	-2.08	6	Garoua		-45.8	-6.71	4	
Buea	-19.3	-3.69	2	Yaoundé		-0.90	-3.18	6	Garoua		-26.8	-2.82	4	
Buea	-37.9	-6.05	2	Yaoundé	Mar 2013	-3.86	-3.57	1	Garoua		-5.8	-1.61	4	
Muyuka	-20.1	-3.56	2	Yaoundé	May 2013	3.00	-2.50	1	Garoua		-12.1	-1.78	4	
Muyuka	-1.56	-1.29	2	Yaoundé	May 2013	25.6	2.33	1	Garoua		1.3	-0.65	4	
Limbe	-5.07	-2.61	2	Yaoundé	May 2013	-36.0	-7.27	1	Garoua		-36.6	-5.64	4	
Limbe	7.89	-0.60	2	Yaoundé	Apr 2013	11.0	-0.44	1	Garoua		-34.0	-4.84	4	
Limbe	-22.4	-3.54	2	Yaoundé	Apr 2013	9.81	-3.99	1	Garoua		-22.5	-3.86	4	
Ndjamena	-22.5	-4.10	8	Yaoundé	Apr 2013	-1.03	-2.62	1	Garoua		-22	-4.17	4	
Diamare	-30.6	-4.20	8	Debunsc		-5.50	-3.03	6	Garoua		-34.7	-4.86	4	
Bakingele	-15.0	-3.02	5	Debunsc		-7.50	-2.94	6	Garoua		-30.5	-4.83	4	
Idenau	-25.4	-3.58	5	Debunsc		-3.60	-2.99	6	Garoua		-3.8	-1.72	4	
Idenau	May 2013	5.13	-2.61	1	Debunsc		-9.10	-3.28	6	Garoua		-25.8	-4.50	4
Brasseries	-24.1	-3.70	5	Debunsc		-16.3	-4.17	6	Garoua		-10.3	-1.57	4	
Upper farm	-27.7	-5.00	5	Debunsc		-20.3	-4.37	6	Garoua		-16.1	-3.51	4	
Foret	-22.8	-4.22	5	Debunsc		-9.5	-3.49	6	Garoua		-39.3	-6.47	4	
Foret SW	-34.7	-5.60	5	Debunsc		-18.7	-4.33	6	Garoua		-11.8	-1.84	4	
Station VHF	-46.5	-6.90	5	Debunsc		-16.0	-3.85	6	Garoua		-12.8	-3.42	4	
Vsant Nord	-44.6	-7.06	5	Debunsc		-12.9	-4.08	6	Garoua		-36.3	-6.45	4	
Foret Buea	-39.7	-6.57	5	Banguem		1.90	-1.95	6	Garoua	Oct 2012	21.6	-1.18	1	
SW BP	-44.6	-7.60	5	Banguem		13.4	-0.56	6	Garoua	Aug 2012	-0.69	-2.02	1	
Bottle Peak	-55.4	-8.02	5	Banguem	May 2013	-19.4	-5.76	1	Benakum	Apr 2013	35.6	3.85	1	
Summit BP	-60.9	-8.69	5	Mt. Man		0.60	-2.20	6	Mbomba	Apr 2013	6.70	-2.48	1	
Summit BP	-64.6	-9.52	5	Mt. Man	Apr 2013	-7.87	-4.65	1	Meigang	Apr 2013	24.2	-0.94	1	
Yaoundé	-58.6	-9.32	6	M. Darlé		17.6	1.13	6	Bertoua	Apr 2013	6.3	-1.53	1	
Yaoundé	0.50	-2.24	6	Garoua		-46.2	-6.37	4	Bamenda	Sep 1986	10.4	-4.00	9	
Yaoundé	-1.40	-2.24	6	Garoua		-37.8	-5.22	4	Bamenda	Apr 2013	21.5	0.16	1	
Yaoundé	-1.00	-2.13	6	Garoua		-27.3	-3.48	4	Ngdéré	Jun 2013	17.8	-0.37	1	
Yaoundé	-5.40	-2.73	6	Garoua		-73.8	-8.46	4	P. Joss	1988-1999	-18.8	-3.85	7	

*I, this study; 2, Ako (2011); 3, Njitchoua and Ngounou Ngatcha (1997); 4, Njitchoua et al. (1995); 5, Gouffantini et al. (2001); 6, Garcin et al. (2012); 7, Mbom and Travi (1994); 8, IAEA (1981, 1991, 2007); 9, Kusakabe et al. (1989).*

line (CMWL). However, the data they used did not cover the entire Cameroonian territory. Consequently, this study attempts to establish a more representative CMWL for Cameroon by using data that covers more territory. Accordingly, in addition to the data obtained in this work, we incorporated literature data (Fontes and Olivry, 1976; Njitchoua *et al.*, 1995; Njitchoua and Ngounou Ngatcha, 1997; Gonfiantini *et al.*, 2001; Ako *et al.*, 2009; Ako, 2011; Garcin *et al.*, 2012). Because the areas are climatologically similar, the weighted averages of the isotopic ratios from Ndjamena in Chad (IAEA, 2007) and data from Mbonu and Travir (1994) in the Joss Plateau of Nigeria were used to supplement the limited data from the northern semi-arid region and the Adamaua Plateau (where we had only 2 samples), respectively.

Ninetyeight data points (Tab. 2) were used to construct the new CMWL with a least squares line equation  $\delta D = 8.6\delta^{18}O + 15.2$  (Fig. 3). The correlation coefficient ( $r^2 = 0.88$ ) reveals a good fit; the narrow 95% confidence band delineated by dotted lines suggests that the statistical parameters of the population (mean, standard deviation, variance) are well constrained by the sampled data. The intercept and slope values of the CWML are slightly different from those reported by Garcin *et al.* (2012), reflecting a better territorial repartition of the samples. The slope, which is slightly greater than 8 (8.6), indicates that the rainfall events mostly occur under slight isotopic disequilibrium with a 5.2‰ excess in  $^2H$  and a 0.6‰ depletion in oxygen-18 compared to the fully equilibrated moisture-condensate fractionation. The factor responsible for the observed disequilibrium may be the mixing of the oceanic-derived air masses with the continentally derived air winds that integrate the southern high evapotranspiration-dominated air masses and the dry derived air masses originating from the Sahara desert.

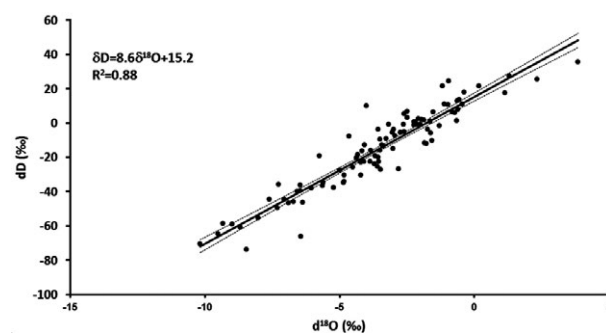
### The intra-lake isotopic variation

To describe the intra-lake variation, the  $\delta^{18}O$  and  $\delta D$  values for the three (for lakes >50 m deep) and two (for lakes <50 m deep) sampled depths were plotted *versus* the depth (Fig. 4). The graphs reveal the following: i) all of the lakes except Lake Oku (Fig. 4 c,d) and, to a lesser extent, lakes Barombi Mbo (SW-LD) and Baledjam Marbuwi (B-m) in the Ad-LD, are isotopically stratified. They exhibit at least 2 distinct layers; ii) irrespective of their location, even lakes <15 m deep are stratified contrary to the observation that shallow lakes and swamps generally have homogeneous water columns (Gonfiantini, 1986). The epilimnia are generally more enriched in 18-oxygen than the hypolimnetic waters. Compared to the other areas, the  $\delta^{18}O$  profiles of the CVL lakes have the same shapes as the profiles of the water found in Saharan dunes (Coplen *et al.*, 2001). However, when compared to the  $\delta^{18}O$  profiles of lakes Malawi and Tanganyika (Gonfi-

antini, 1986), which are also isotopically stratified, the latter show reversed isotopic profiles.

The isotopic structure shown above might not thoroughly depict the actual distribution of the water columns because only three depths were sampled, as mentioned above. Consequently, the isotopic gradient (*isotopicline*) may be positioned at a shallower depth. Furthermore, due to the limited number of sampled depths, the sub-layers and micro-layers (*e.g.*, lakes Nyos and Monoun) may not appear. Therefore, the profiles in Fig. 4 may describe the overall (bulk) structure of the water columns, particularly regarding their stratification. The chemical or thermal stratification in the lakes is generally driven by a density gradient. In subtropical areas, almost all of the deep lakes are thermally stratified due to the energy contributed by solar radiation. Lakes like lakes Nyos and Monoun are both thermally and chemically permanently stratified (Evans *et al.*, 1994; Kling *et al.*, 1987, and other references therein). For instance, whether the observed isotopic stratification is seasonal or permanent remains a subject of investigation. Based on general observations, the isotopic characteristics of the lakes are discussed from the SW-LD, the Bd-LD, then the Ad-LD.

The SW-LD comprises five lakes. However, Fig. 4 a,b exhibits four profiles because at Lake Barombi Koto (less than 5 m deep, see Tab. 1), only one sample was collected from the middle depths. Of the 4 lakes, Barombi Mbo (BM) is the most enriched in heavy isotopes, followed by Lake Manenguba Female (Mf), which is more enriched in  $^{18}O$  than Lake Debunsha (Db); the latter is comparatively more enriched in  $^2H$ . Lake Manenguba male (Mm) has the lightest waters and exhibits the strongest isotopic gradient. In addition, the  $\delta^{18}O$  and  $\delta D$  in the Mm do not co-vary throughout the depth range: while the  $\delta^{18}O$  profile is marked by a positive isotopic gradient from the surface to approximately 45 m (remember that the depth might not be the exact location of the gradient, as explained above)



**Fig. 3.** The Cameroon Meteoric Water Line (CMWL) derived from 97 isotopic ratios distributed across the Cameroonian territory. The dotted lines delineate the 95% confidence band.

Tab. 3. Isotopic composition of groundwater, rivers and lakes.

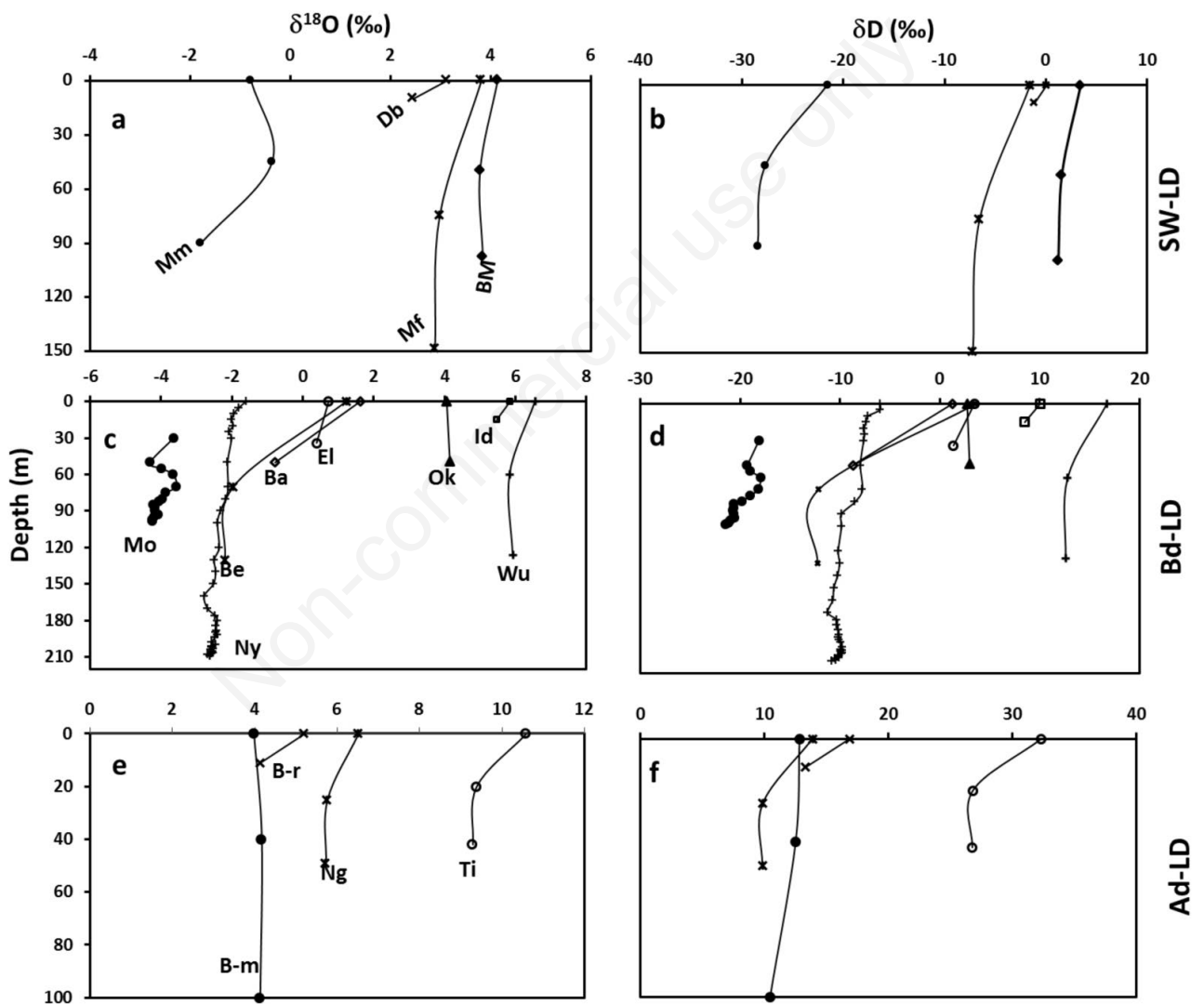
Lakes District	Ground and surface water				Lakes						
	Site	Date	$\delta D$	$\delta^{18}O$	Name	Date	Depth (m)	$\delta D$	$\delta^{18}O$	Mean	Mean
BM	Db Gw	May 2013	-2.8	1.0	Barombi Koto	Apr 2013	0	9.3	5.5		
	BK Gw	Apr 2013	-14.3	0.1	Debunsha	May 2013	0	0.1	3.1		
	BM Gw	Nov 2012	-16.0	-0.3	Debunsha	May 2013	10	-1.1	2.4		2.8
	BM Vi	Nov 2012	-8.2	1.3	Manenguba Male		0	-21.5	-0.8		
	Inlet 1	Nov 2012	-15.0	-0.3	Manenguba Male		45	-27.7	-0.4		
	Inlet 2	Nov 2012	-15.1	-0.1	Manenguba Male		90	-28.4	-1.8		-1.0
	Inlet 3	Nov 2012	-15.2	-0.5	Manenguba Female	May 2012	0	-1.5	3.8		
	Inlet 3	Apr 2012	-7.4	1.3	Manenguba Female		75	-6.5	3.0		
	Inlet 1	Apr 2012	-9.4	1.2	Manenguba Female		149	-7.2	2.9		3.2
	Inlet 2	Apr 2012	-9.9	1.0	Manenguba Female		0	3.4	4.1		
SW-LD	Ndi.Gw	Apr 2012	-22.0	-0.2	Barombi Mbo		50	1.6	3.8		
	Tap 1	Apr 2012	-27.8	-0.9	Barombi Mbo		98	1.2	3.8		3.9
	Tap 2	Apr 2012	-28.1	-0.9	Baleng	Apr 2013	0	1.2	1.6		
	Inlet-a	May 2012	-29.5	-2.3	Baleng	Apr 2013	50	-8.7	-0.8		0.4
	Inlet-b	May 2012	-26.2	-0.9	Benakuma	Apr 2013	0	3.6	1.2		
	Evam1	May 2012	-24.1	-0.4	Benakuma		70	-12.2	-2.0		
	Evam2	May 2012	-23.9	-0.6	Benakuma		130	-12.3	-2.2		-1.0
	Mboue	May 2012	-26.8	-0.9	Oku	May 2013	0	2.7	4.1		
	Bal. Gw	Apr 2013	-17.7	-0.2	Oku		49	3.0	4.1		4.1
	Bef. Gw	Apr 2013	-9.3	0.6	Idjagham	May 2013	0	10.0	5.8		
Bd-LD	Ok. Gw	May 2013	-26.0	-1.9	Idjagham		15	8.5	5.5		5.7
	Idj. Gw	May 2013	-10.3	0.8	Elum	Apr 2013	0	3.4	0.7		
	Mo.	Mar 2013	-18.4	-2.3	Elum		34	1.4	0.4		0.6
	Mo. Gw	Mar 2013	-20.4	-2.9	Wum	Apr 2012	0	16.7	6.5		
	Riv.Pan	Mar 2013	-11.3	-1.9	Wum		60	12.8	5.8		
	Tiz.Gw	Apr 2013	-13.7	0.3	Wum		126	12.6	5.9		6.1
	Dib.Gw	Apr 2013	-16.0	0.0	Ngaoundaba	Apr 2012	0	13.9	6.5		
					Ngaoundaba		25	9.9	5.8		
					Ngaoundaba		49	9.8	5.7		6.0
					Tizon	Apr 2012	0	32.3	10.6		
Ad-LD				Tizon		20	26.8	9.4			
				Tizon		42	26.8	9.3		9.8	
				Baledjam Marbuwi		0	12.8	4.0			
				Baledjam Marbuwi		40	12.5	4.2			
				Baledjam Marbuwi		100	10.4	4.1		4.1	
				Baledjam (Ranch)		0	16.9	5.2			
				Baledjam (Ranch)		11	13.3	4.1		15.1	4.7

Db, Lake Debunsha; BK, Lake Barombi Koto; BM, Lake Barombi Mbo; Mf, Lake Manenguba Female; SW-LD, South-west Lake District; Bd-LD, Bamenda Lake District; Ad-LD, Adamau Lake District.



and then reverses toward the bottom, the  $\delta D$  exhibits a negative gradient downwards to the same depth before remaining stable, implying that the two elements are decoupled. Although differences in the isotopic distribution along lake water columns are common (Gonfiantini, 1986), the  $\delta^{18}O$ - $\delta D$  decoupling might be a rare phenomenon. The Manenguba twin lakes (Mf and the Mm) are located approximately 200 m apart on the same caldera (Fig. 5). Therefore, they experience the same meteorological conditions. Consequently, why are they so isotopically

different? A clear-cut answer to that question may be difficult to obtain. However, these remarkable isotopic differences might be due to their morphometry (Jones and Imbers, 2009) and/or their biotopes (Pettit, 1936). First, the relative enrichment in Mf could occur because the lake is large and open. Therefore, it may undergo more evaporation, which occurs primarily due to wind (Birge, 1916; Garwood *et al.*, 1985), compared to the Mm, which is located 20 m lower (Mm 1900 m asl, as against 1920 m asl for the Mf) in a deep crater with high walls. That setting

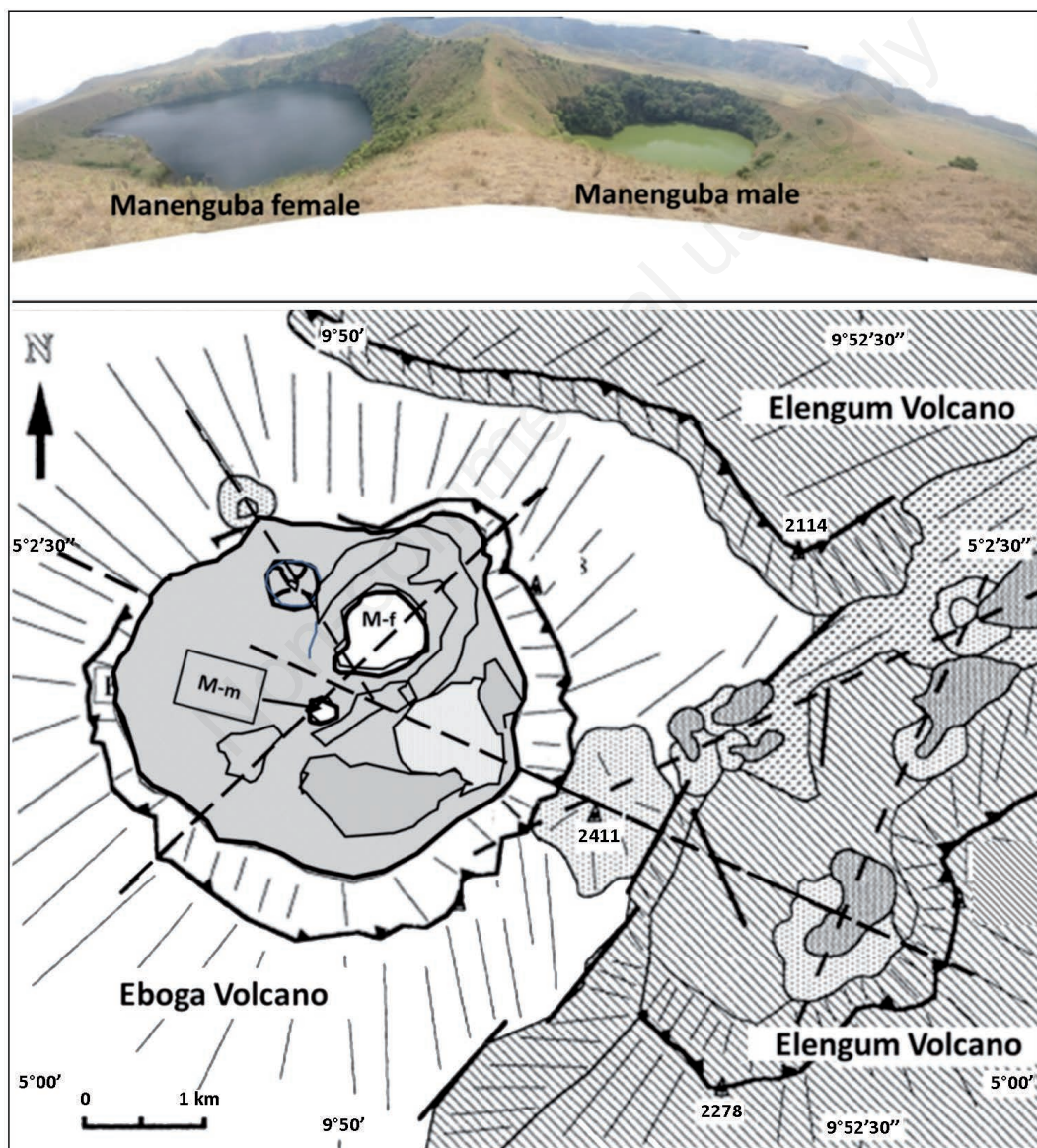


**Fig. 4.**  $\delta^{18}O$  and  $\delta D$  profiles of the lakes. a,b) South-West lake district (SW-LD); Mm, Manenguba male; Mf, Manenguba female; Db, Debunsha; BM, Barombi Mbo. c,d) Bamenda lake district (Bd-LD); Mo, Monoun; Ny, Nyos; Be, Benakuma; Ba, Baleng; El, Elum; Ok, Oku; Id, Idjagham; Wu, Wum. e,f) Adamaua lake district (Ad-LD); B-m, Baledjam Marbuwi; B-r, Baledjam ranch; Ng, Ngaoundaba; Ti, Tizon. All the lakes, except Oku, Baledjam and Barombi Mbo are more or less stratified with heavy isotopes enriched surface waters. Even shallow lakes (>15m) are stratified. The largest gradient between surface and bottom waters is observed in Lake Benakuma. Lake Baledjam Marbuwi is distinguished by a hypolimnion being more enriched than epilimnion. Note the  $\delta^{18}O$  and  $\delta D$  decoupling in the Mm.

may shield the latter from the wind. Second, the two lakes have different colors (Fig. 5): the Mf is dark, while Mm is green. The green coloration displayed by volcanic lakes is often attributed to chemical reactions triggered by the volcanic gas activity that drives nutrient-rich water toward the surface (Tiwu Nuwa Muri Koo Fai Lake, Keli Mutu volcano, Indonesia) and/or to the high nutrients contents and/or algal growth (Morris and Hargreaves, 1997). As the Manenguba area is not volcanically active, the green color is attributed to the microbial content of the lake. Depending on the metabolic cycles and paths, the micro-organic

metabolic processes (e.g., respiration and photosynthesis) can induce a negative shift in the  $^{18}\text{O}$  composition of a lake (Pushkar *et al.*, 2008). This observation may cause the isotopic decoupling and change the isotopic fingerprint of the twin lakes (Mm and Mf).

Lake Barombi Mbo (BM), which is located approximately 60 km wing fly to the south-west of Mount Manenguba at approximately 301 m asl, is the largest crater lake in Cameroon (Chako Tchamabé *et al.*, 2013). The lake is approximately 100 m deep. Similar to Lake Baledjam Marbuwi in the Ad-LD (Fig. 4 e,f), its  $\delta^{18}\text{O}$  and  $\delta\text{D}$



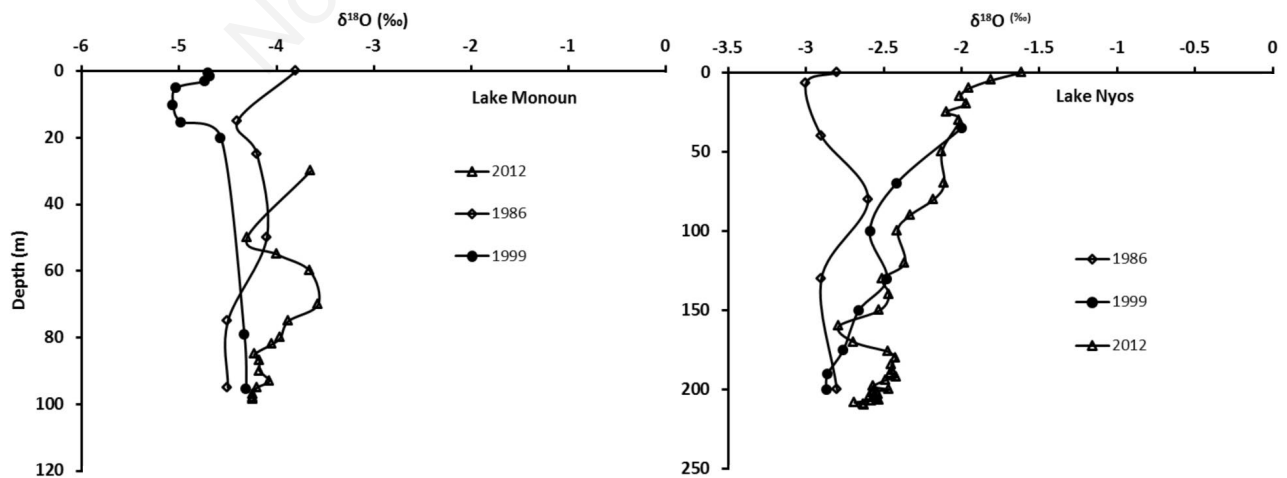
**Fig. 5.** View of the twin lakes Manenguba and the Manenguba caldera’ geological setting composed of two structures: the Elengum volcano and the Eboga volcano, the most recent formed (0.51 Ma) hosts the caldera. The female Lake is the largest and the male has a green color and is hosted in a deep crater with high walls.

profiles indicate that its water column is nearly isotopically homogeneous (Fig. 4 a,b) compared to lakes Benakuma (Be) and Wum (Wu) in the Bd-LD or the Manenguba twin lakes, which are similar in depth (Tab. 1). Currently, whether the BM always features such homogenous water column remains unknown. However, the observed isotopic distribution may be due to the overturn that Lake Barombi Mbo underwent several months before the survey (IRGM, *internal report*).

Finally, Lake Debunsha (Db) is located a few hundred meters from the Atlantic Ocean at approximately 54 m *asl*. Due to the approximately 12,000 mm/year of rainfall it receives (the precipitation amount is almost equal to its depth=13 m), its water column was expected to be homogenous and isotopically similar to the precipitations (means  $\delta^{18}\text{O}=-3.6\text{‰}$ ,  $\delta\text{D}=-11.9\text{‰}$ ). Surprisingly, the lake is clearly stratified, implying that the high humidity and the expected rapid recycling of the isotopes due to the frequent precipitation near the coast exert a minimal effect on the isotopic composition of that lake. Instead, the lake seems to undergo noticeable isotopic fractionation, especially the surface water ( $\delta^{18}\text{O}=3.1\text{‰}$ ,  $\delta\text{D}=0.1\text{‰}$ ) due to the wind-driven evaporation.

The Bamenda Lake District (Bd-LD) is located to the North of the SW-LD. As noted above, all of the lakes in that LD except Lake Oku (Ok; Fig. 4 c,d), exhibit weak to relatively strong isotopic stratifications. Lake Oku is located in the highlands at approximately 2227 m *asl*. This area is characterized by small daily variations in temperature (Letouzey, 1985) that, when combined with the size of the lake (52 m deep only, see Tab. 1), may help maintain the homogenous water column structure through internal mix-

ing. Lake Benakuma, (Be), (Fig. 4 c,d), (the third deepest lake of the CVL after lakes Nyos and Manenguba Female) and to a lesser extent Lake Baleng show the strongest isotopic gradients among the CVL lakes. That relatively strong stratification may limit the material transport between the epilimnion and the hypolimnion, which is depleted in heavy isotopes compared to the present-day rainfall ( $\delta^{18}\text{O}=3.8\text{‰}$ ,  $\delta\text{D}=35.6\text{‰}$ ); therefore, it is thought that, the water of interest was precipitated and/or probably recharged under colder conditions. Due to their behavior, the gassy lakes (Nyos and Monoun) are of particular significance. Their isotopic profiles (Fig. 6) reveal that their water column includes several layers that are separated by weak isotopic gradients. They also have the lowest isotopic values, which are similar to that of the local ground. The  $\text{CO}_2$ -rich bottom waters are even more depleted due to the isotopic fractionation between the  $\text{CO}_2$  and the water molecules, which should enrich the dissolved carbon dioxide in 18-oxygen at the expense of the water; this behavior was predicted by a fractionation factor  $\alpha_{\text{CO}_2\text{-H}_2\text{O}}=ca. 1.041$  (Bottinga and Craig, 1968; Brenninkmeijer *et al.*, 1983). The effects of this process might be apparent in all of the lakes with a large,  $\text{CO}_2$ -rich gas reservoir. Therefore, the presence of isotopically light deep waters might be used as indirect evidence for the presence of a gas reservoir in volcanic lakes. Fig. 6 also shows the changes in the isotopic profiles of lakes Nyos and Monoun over time. At Lake Nyos, the isotopic composition of the 1986 profile, which was measured several months after the gas burst, falls between the 1999 and the 2010 profiles. Between 1986 and 1999, the surface waters became isotopically lighter before the changes reversed toward a heavier trend from 1999 to 2010. That surface evo-



**Fig. 6.**  $\delta^{18}\text{O}$  profiles of lakes Nyos and Monoun (2012, 2010 and 1999 data from Sasaki *et al.* 2012, *personal communication*; Nagao *et al.*, 2010 and Kusakabe *et al.*, 1989, respectively). Contrary to the TDS that increase from the surface to the bottom,  $\delta^{18}\text{O}$  instead shows a negative gradient.  $\text{CO}_2$ -rich bottom waters are the most depleted assumedly because of the isotopic fractionation between  $\text{CO}_2$  and water.

lution may simply reflect the environmental conditions. At the middle depths, the 1986 profile features a layer enriched in heavy isotopes that most likely represents a mixture of the degassed water from the gas exhalation and the subsided evaporated rainwater. The homogenous water column structure of the 1999 profile indicates that, the lake became more enriched in 18-oxygen between 1986 and 1999. Afterwards, a different water column structure characterized by two anomalous layers at middle depths developed. That change may reflect the on-going gas removal (Halbwachs *et al.*, 2004; Kusakabe *et al.*, 2008; Issa *et al.*, 2013) and the ground water inflow. In contrast to the surface and middle depths, which alternately experienced isotopic enrichment and depletion during the reference period, the deepest waters (*monimolimnion*) became heavier over time. At Lake Monoun, the 1999 profile exhibits the lowest  $\delta^{18}\text{O}$  values. Although the surface layer thickness (approximately 18 m) was maintained throughout the observation period, reflecting the constant volume of water inflow (*e.g.*, from River Panke), it became more enriched in  $\delta^{18}\text{O}$  due to variations in the inflow/evaporation ratio. Notably, an anomalous isotope-concentration layer was developed at between 60 m and 85 m, suggesting the presence of ground water inflow with a comparatively positive isotopic composition. Concurrently, the isotopic composition of the bottom water did not change much after 1986.

The evolutionary trend discussed above shows that the anomalies created by the inflows could be traced, and their origin could be constrained. The gassy lake water is structurally controlled by the total dissolved solids (TDS), which increase from the surface toward the bottom (Evans *et al.*, 1994; Kusakabe *et al.*, 2008; Issa *et al.*, 2013). In contrast, the  $\delta^{18}\text{O}$  concentrations decrease toward the bottom, implying that the TDS do not influence the  $\delta^{18}\text{O}$  concentrations or that the isotopic fractionation between the water and dissolved  $\text{CO}_2$  exerts greater control than the dissolved salt (Gat, 1995). If the latter assertion is true, the gas accumulation should be traceable through the 18-oxygen.

Finally, the lakes in the Adama Lake District (Ad-LD) are the heaviest with average  $\delta^{18}\text{O}$  and  $\delta\text{D}$  values above 6.1‰ and 16.7‰, respectively (Fig. 4 e,f). Lake Tizon exhibits the highest isotopic ratios ( $\delta^{18}\text{O}$ =9.8‰ and  $\delta\text{D}$ =28.6‰, Tab. 3). As mentioned above, Lake Baledjam Marbuwi (B-m) is the only one to display  $\delta^{18}\text{O}$  enriched bottom water relative to the surface water.

### **Effect of mixing with local water bodies (precipitations, rivers and ground water)**

#### ***Contribution of the precipitations to the isotopic composition of the lakes***

To qualitatively evaluate and constrain the isotopic contribution of the present day precipitation to the isotopic compositions of the lakes, we plotted the average isotopic

composition of the lakes and the mean composition of the precipitation from each region (Fig. 7). The local evaporative line (LEL) of each Lake District and the CMWL are also shown.

For a given area, the isotopic composition of the source rain for the evaporating water bodies could be constrained by back extrapolating the local evaporative line to intersect with the local meteoric line; the intersection should provide the isotopic composition of the source rain (Gat, 1995; Kebede, 2004; Henderson and Shuman, 2010). For the Ad-LD, Fig. 7 indicates that the predicted isotopic composition of the source rain is close to that of the measured rain. In contrast, the LELs of the SW-LD and NW-LD intersect quite far from the measured isotopic values of the source rain in the two regions for unknown reasons. To overcome these mismatches, an alternative method was used. The upper and lower limits of the  $\delta^{18}\text{O}$  values from a given group were horizontally back-projected onto the CMWL and vertically projected on the abscissa axis. Consequently, instead of a point, intervals containing the theoretical values of the  $\delta^{18}\text{O}$  composition of the source rain are obtained, as marked by the double arrows (Fig. 7). Similarly, the corresponding  $\delta\text{D}$  values could be obtained by horizontally projecting the intersections on the Y axis. The results indicate that the averaged  $\delta^{18}\text{O}$  values of the source precipitation in the SW- and Bd-LDs fall within the predicted intervals (horizontal double arrows continuous line (black) and the dash dot line, respectively). Therefore, the lakes in these two LDs contain a significant amount of minimally evaporated water with an isotopic composition similar to that of the present day precipitation. Conversely, the mean average  $\delta^{18}\text{O}$  of precipitation in the Ad-LD falls slightly out of the expected interval (double arrow dash line) while remaining close enough to the lower limit value; therefore, compared to those in the southern region, the Ad-LD lakes may contain proportionally less water with the present day isotopic signature of the rainfall, and/or they have undergone more evaporation based on their geographical locations.

#### ***Mixing of lakes water with rivers and ground waters***

To assess the degree of mixing with the local ground water, we plotted (Fig. 8) the isotopic compositions of the lakes with the averaged isotopic compositions of the local groundwater and inlet/outlet rivers (Darling *et al.*, 1996; Ayenew, 1998; Chernet, 1998; McKenzie *et al.*, 2001; Kebede *et al.*, 2002; Delalande *et al.*, 2008; Gat, 2010).

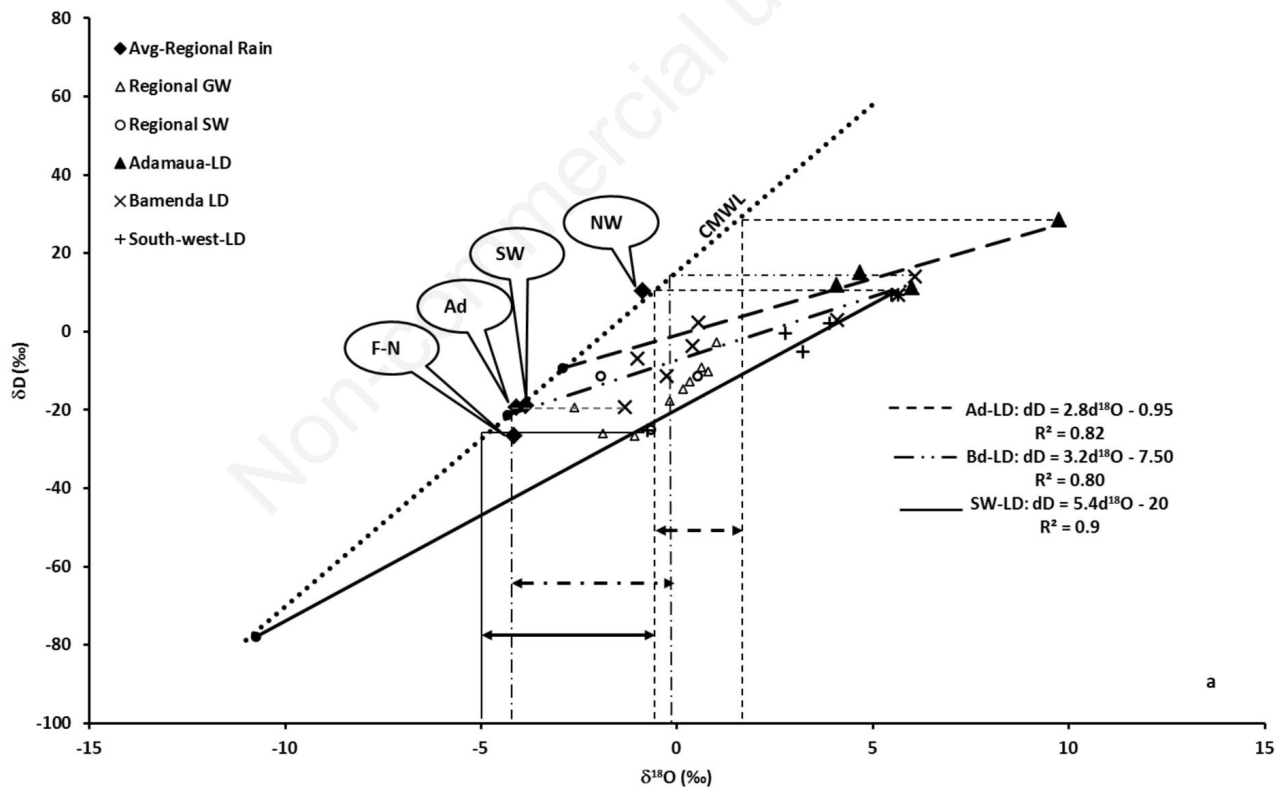
The lakes in the SW-LD are plotted slightly away from the ground water, implying they had limited connectivity with the local hydrogeological systems and/or that they may be hydraulically connected to several different aquifer systems. For example, the isotopic compositions of the Lake Manenguba male bottom water and the inlet spring water (Mm inlet:  $\delta^{18}\text{O}$ =-2.3‰ and  $\delta\text{D}$ =-29.5‰; Tab. 3) are

similar, indicating that the two water bodies most likely tap the same aquifer system containing heavy isotope-depleted water that is recharged under low-temperature and high-humidity conditions. The surface water of this lake seems to result from a mixture of rain ( $\delta^{18}\text{O}=-5.8\text{‰}$ , Tab. 2), ground water ( $\delta^{18}\text{O}=-2.3\text{‰}$ , Tab. 3) and water from a different aquifer system with  $\delta^{18}\text{O}=-0.9\text{‰}$ , which is comparable to the isotopic composition of the Mf inlet spring (Tab. 3). The isotopic composition of Mf is different from its inlet and all of the sampled ground water in the region. Its water had undergone significant evaporation (as compared to the Mm) due to its location, as explained above.

The water from BM is remarkably enriched in heavy isotopes relative to its inlets (inlet 1, 2 and 3; Tab. 3):  $\delta^{18}\text{O}=0.4\text{‰}$ ,  $\delta\text{D}=-12.0\text{‰}$  and the regional ground water (BK Gw, BM Gw, BM Vi, B) with mean isotopic composition of  $\delta^{18}\text{O}=0.3\text{‰}$ ,  $\delta\text{D}=-12.8\text{‰}$  (Tab. 3). Similar to Mf and Db, the difference in the isotopic ratio between the

lake and the surface and ground water inflows indicates that the contribution of the emissary toward the isotopic composition of the lake is limited. In fact, the BM supplies drinking water to the neighboring city of Kumba. Consequently, the majority of the water contributed by the inlets might be extracted for that purpose.

Except for lakes Wum, Idjagham and Oku, most of the lakes in the Bd-LD cluster have ground and surface water that indicates extensive hydraulic connectivity. Through a slight extrapolation, Lake Oku seems to tap water from an aquifer with a different isotopic composition from that of the sampled water ( $\delta\text{D}=-26.0\text{‰}$  and  $\delta^{18}\text{O}=-1.9\text{‰}$ ), making it significantly different from that of the lake ( $\delta^{18}\text{O}=4.1\text{‰}$ ,  $\delta\text{D}=2.8\text{‰}$ ) (Tab. 3). This situation is also applicable to lakes Wum and Idjagham, which have isotopic compositions that are significantly different from that of the sampled ground waters with isotopic ratios of  $\delta\text{D}=-9.3\text{‰}$  and  $\delta^{18}\text{O}=0.6\text{‰}$ ; and  $\delta\text{D}=-10.3\text{‰}$  and  $\delta^{18}\text{O}=0.8\text{‰}$ , respectively.



**Fig. 7.** Plot of regional isotopic compositions of precipitation and the mean isotopic composition of the lakes. In order to constrain the isotopic signature of the source rain in each lake district (LD), the upper and lower limits of each local evaporative line (LEL) was horizontally back-projected to the Cameroon Meteoric Water Line (CMWL); the intersections were then vertically projected on the abscissa axis to define an interval which may contain the  $\delta^{18}\text{O}$  value of the source rain. In a similar way, an interval could be defined by projecting the intersection on the Y-axis to obtain the corresponding  $\delta\text{D}$ . The regional rain of the SW-LD and Bd-LD fall within the expected interval defined by the horizontal solid double arrows and the horizontal dash-dotted double arrows. However, isotopic signature of rains in the Ad-LD falls slightly out of the interval delineated by the horizontal dashed double arrows. SW, stream water; GW, ground water. Regional rain (circled): N-W, North-West (Bd-LD); S-W, South-West (SW-LD; Ad, Adamaua (Ad-LD); F-N, Far-North.

Finally, the lakes in the Ad-LD are the ones that are most different from the local ground and surface waters, implying that their interactions are very limited. However, it is possible the climate-driven isotopic fractionation processes (Drever, 2002 and other references therein) mask the lake-groundwater-surface water relationship in that LD. In addition to the mixing effect with the local water bodies, another important factor can account for the present isotopic composition of the lakes. Long-term evaporation-mixing-subsidence processes may also shape the isotopic distribution along the water columns. In fact, when strong stratification allows material transport between layers, the temperature variations between day and night promote downward circulation (Rowe *et al.*, 1992) for the water from the isotopically more positive epilimnion toward the hypolimnetic water strata during the night. In the long-term, depending on the residence time, the subsidence will isotopically enrich the lake compared to the local ground water. This process could be envisaged for BM, which is minimally or not stratified.

### Spatial variation in isotopic composition and evaluation of the control parameters

#### Spatial variation

To evaluate the influence of the geographical setting on the isotopic composition of the lakes, the surface, middle and bottom isotopic ratios were averaged to yield a representative isotopic composition for each lake (Tab. 3). The

values were plotted in the conventional  $\delta D$ - $\delta^{18}O$  space with the CMWL and presented in Fig. 9. Because we are evaluating volcanic lakes, which may connect with the hydrothermal systems, the fields for metamorphic and magmatic waters (Taylor, 1979; Giggenbach, 1992) have also been included. The lakes plot to the right of the CMWL along a least square line  $\delta D = 3.8 \delta^{18}O - 10.2$ ,  $r^2 = 0.82$  (designated hereafter as Lakes Water Line LWL) indicating enrichment in heavy isotopes. The remarkable displacement in the  $\delta O^{18}$ - $\delta D$  space of the African lakes compared to lakes in other parts of the world was previously observed during the early surveys of stable isotope variations in the hydrological cycle (Gat, 1995). The slope of the LWL (3.8) falls within the predicted interval (3.5-6.5) for open, evaporating water bodies (Dincer, 1968; Kebede, 2004). However, it is much lower than that for Ethiopian lakes (5.2 to 6.4) and other lakes in Eastern (Turkana) and or Central Africa, such as Lake Chad (Fontes *et al.*, 1970). In contrast to the northward impoverishment of the rain in heavy isotopes due to the latitude effect (Dansgaard, 1964; Gat, 2010), the lakes became more enriched further inland.

Based on the LELs, (Fig. 7) a theoretical evaluation of the latitudinal evaporative effect on each group of lakes is attempted. Their slopes decrease from 5.4 in the SW-LD to 2.9 in Ad-LD while passing through the value of 3.3 in the Bd-LD (Fig. 7). The slopes of the two Southern-most LDs fall within the theoretical interval for open evaporating water bodies (Kebede, 2004). In contrast, the value for the Ad-LD falls outside of that range and ap-

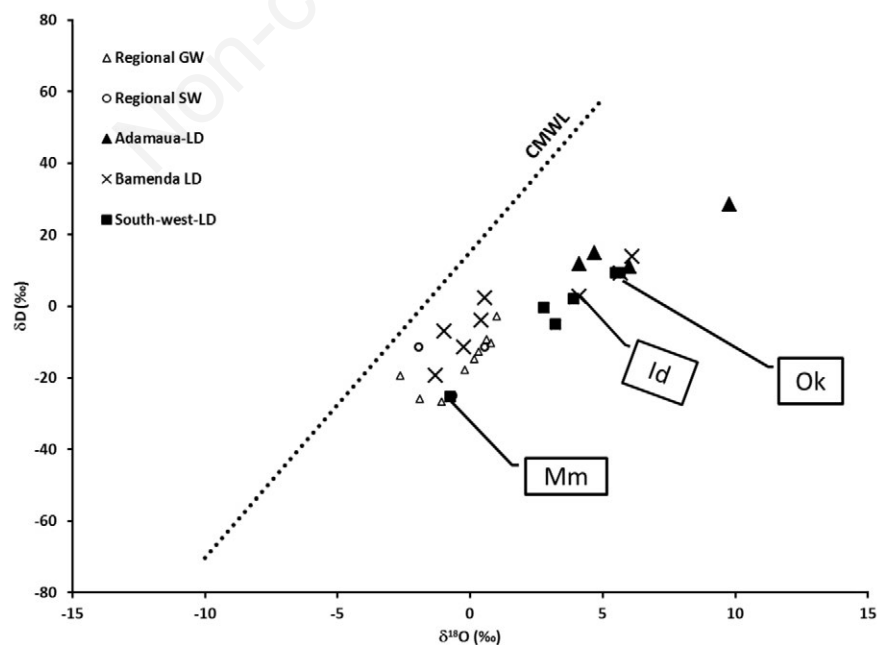


Fig. 8. Plot of the mean isotopic composition of the lakes and that of regional surface and ground water.

proaches that of the high temperature fluids (2.1) (Fig. 9) encountered in arc volcanic regions (Taran *et al.*, 1989; Giggenbach, 1992), suggesting severe evaporation. Because the mean temperature (*ca* 20°C) in the Ad-LD is much lower than that of the southern LDs (24–26°C), this observation may imply that the temperature-dependence isotopic fractionation might be reversed along the CVL. That reversion may be attributed to the varied and marked phyto-climatological settings. The Ad-LD the vegetation cover is pre-savanna (Fig. 2c), and the ambient humidity is low; these conditions may favor kinetic isotopic fractionation throughout the 7 months of the dry season. Conversely, the equilibrium isotopic fractionation might be the dominant process in the two southern LDs, which are characterized by a dense evergreen forest and high humidity throughout the year (Fig. 2c). Therefore, the equilibrium fractionation processes, which often result in abundant rainfall, may significantly deplete the water reservoirs through the amount effect.

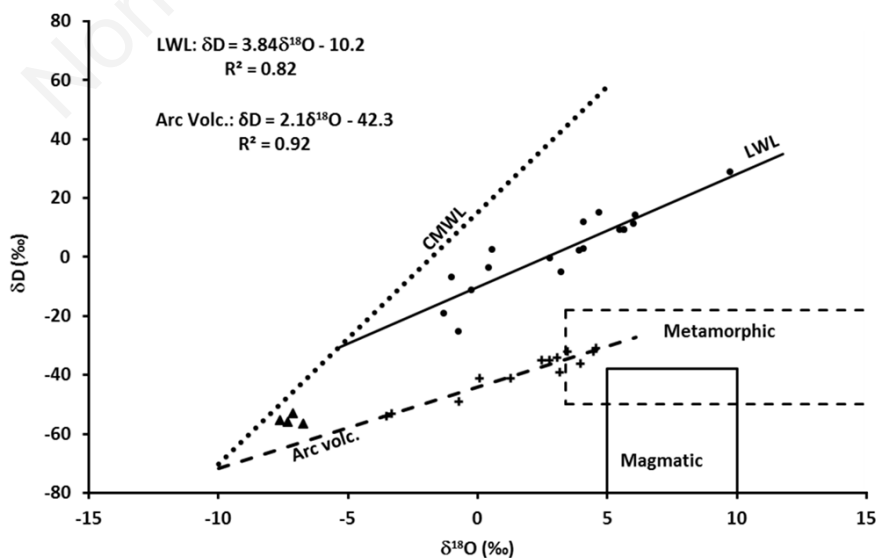
Similar to the increase in the values of the slopes, the d-excess also increases from -20 in the SW-LD to -7.5 in the Bd-LD, reaching -1 in the Ad-LD. This trend reveals the influence of the specific local physical conditions on the oceanic source of the precipitations. The northward increase in the d-excess reflects the decrease in the precipitation/evaporation ratio (Sighomnou, 2004) and the prevailing conditions during the mixing of oceanic and the Saharan derived air masses, which converge to form to ITCZ that controls the seasonality across the country. As indicated above, Lakes Wum, Idjagham and Oku in the Bd-LD plot falls among the lakes of the Ad-LD (Fig. 8), sug-

gesting that in addition to the climatologic controls, the interaction between the lakes and their immediate environments, such as the hydrological setting (Cabassi *et al.*, 2013) and intrinsic parameters (such as size, depth, sheltering from wind, *etc.*), cannot be ignored regarding the isotopic composition of the CVL lakes.

### Quantification of the influence of some parameters

Evaluating the influence of the environmental and/or intrinsic factors on the variations in the water isotopes can lead to a better understanding of the fractionation and control processes. In this study, we selected five parameters to evaluate their effect on the isotopic composition of the CVL lakes: the estimated average temperature, the altitude, the maximum depth, the distance from the coast and the hydrological setting (data from Tab. 1). These parameters have been plotted *versus*  $\delta^{18}\text{O}$  and are displayed in Fig. 10.

The scattering of the points in graphs a, b, c and d is supported by the very weak correlation coefficients ( $0 \leq r^2 \leq 0.15$ ), suggesting that the parameters mentioned above exert a negligible influence on the variations in  $\delta^{18}\text{O}$  variation. However, when evaluated relative to one another, the mean regional temperature and altitude seem to play the smallest role in the isotopic composition of the lakes (Fig. 10 a,b). In contrast, the depth exerts a comparatively greater and discernible influence (Fig. 10c), while the effect of the latitude (distance from the coast) was estimated at +0.6‰/100 km as one moves from the South to the North (Fig. 10d). Although it is not quantifiable, the effect of the hydrological setting is noticeable because the

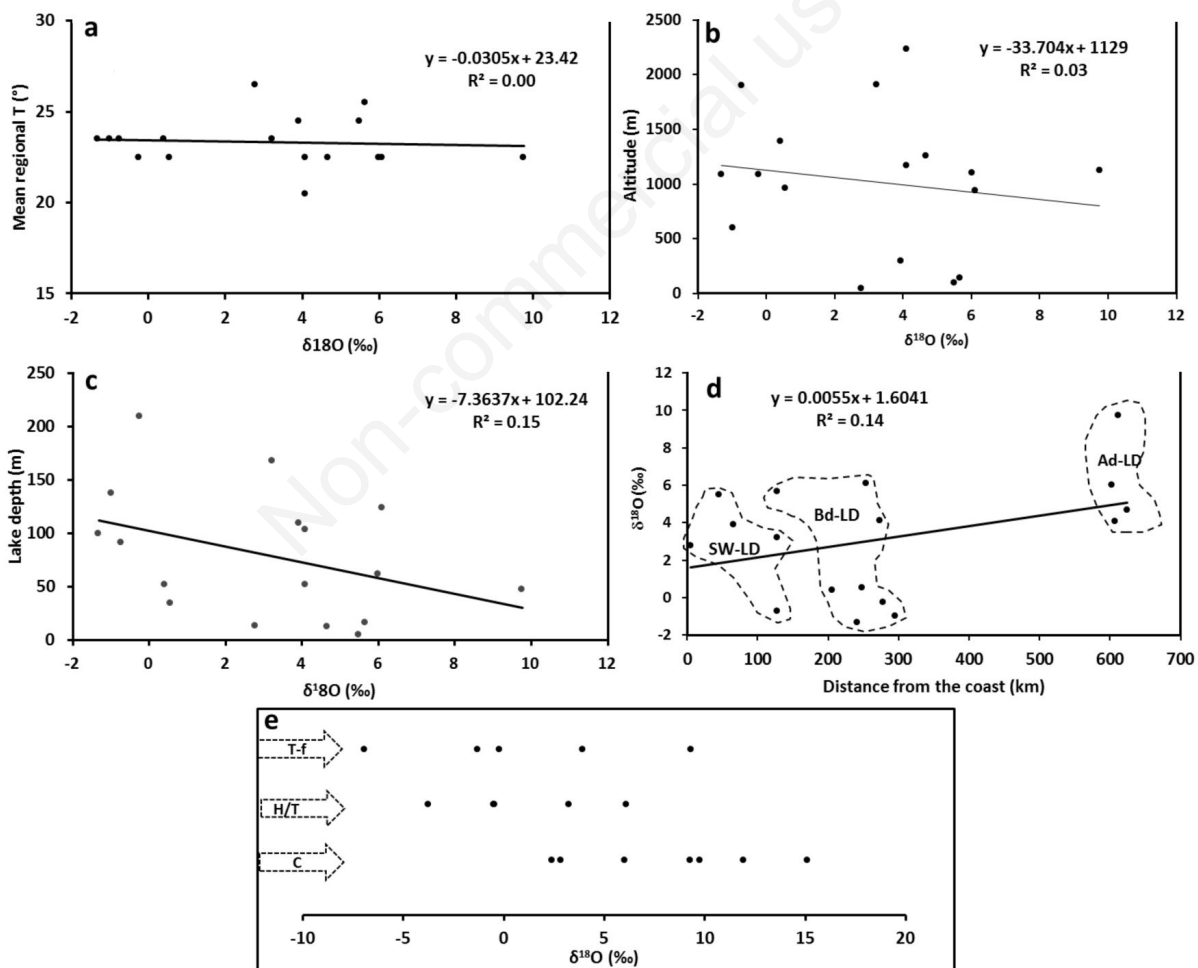


**Fig. 9.** Plot of averaged isotopic composition of the lakes in the  $\delta^{18}\text{O}$ - $\delta\text{D}$  space. Metamorphic and magmatic fields are shown. The evaporative line for high temperature fluids (data from literature, see text) is also displayed.

closed lakes have the most positive isotopic values (e.g., Lake Tizon is the isotopically heaviest lake) compared to the others, particularly the through-flow lakes (Fig. 10e). The latter observation is consistent with how the mixing of the surface inflows, which are generally depleted in heavy isotopes, lowers the isotopic concentration of the lakes in contrast to the closed lakes, which suffer continuous evaporation without replenishment.

Although this study has qualitatively described and attempted to quantify the effects of factors that could explain the actual isotopic signature of the lakes, better data for those factors are necessary to hierarchize their respective effect adequately; the data are regional rather than site-specific. This work indicates that the variations in isotopic content attributed to interactions between a lake and the exogenous waters can be reasonably deciphered and that the origin of the exogenous waters can be identified

if accurate and sufficient data regarding the climatology, geology and the hydrology are available. Although it is complex, the availability of this information should enable qualitative and quantitative evaluations of the evaporative and/or degree of the mixing effect on open water bodies, such as lakes. For example, Henderson and Shuman (2010) proposed a polynomial that integrates the climatological, hydrological geographical settings to explain the isotopic signature of lakes in the western USA. Jones and Imbers (2009) quantified the relationship between the isotopic compositions and size parameters of lakes. In the absence of geothermic heat, evaporation is primarily controlled by the ambient temperature, as well as dependent factors such as latitude, altitude, climate, humidity wind, (Patalas, 1984; Imberger, 1985) and intrinsic parameters. As shown above, deciphering the influence of each given parameter remains possible.



**Fig. 10.** Some possible control parameters vs  $\delta^{18}\text{O}$ . Temperature (a) and the altitude (b) seem not to play a remarkable role in controlling the isotopic composition of the lakes. Depth (c), distance from the coast (d) and the hydrological setting (e) show weak, however, discernible influence.



## CONCLUSIONS

Seventeen lakes, 47% out of the 39 documented volcanic lakes located on the CVL have been characterized isotopically. Irrespective of their location, size parameters or hydrological setting, the CVL lakes were isotopically stratified with heavy isotope-enriched epilimnia compared to hypolimnetic waters. In several lakes, the isotopic gradient between the surface and bottom waters was large, indicating that these lakes might be meromictic. Whether the stratification is permanent or seasonal must still be clarified because, permanent stratification might favor the accumulation of gas (*e.g.*, Lakes Nyos and Monoun). An attempt has been made to qualitatively and quantitatively evaluate the effect of some parameters on the isotopic variation of the lakes. However, none of the parameters play a predominant role. Consequently, without accurate information regarding those parameters, the interplay between the factors may help shape the isotopic composition of the lakes. The present study is a starting point for studying the isotopes in the CVL lakes, providing a much-needed snapshot of their present status to monitor the subsequent changes that might occur in their water columns (Rowe *et al.*, 1992). The variations in the isotopic profiles of Lakes Nyos and Monoun over time suggest that anomalies along the water column can be detected using water isotopes. Beyond the studied lakes, this work has contributed knowledge regarding the isotopic characteristics of the remaining CVL lakes because about half of this group was studied. This knowledge may be useful when establishing a minimal volcanic lakes monitoring/surveillance network.

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