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## Reply to Comment by Terence McCarthy and Marc Humphries on “Highly Contrasted Geochemical Pattern in Sediments of the Okavango Delta, Botswana Driven by Dust Supply, Hydrological Heritage and Biogeochemical Reactions”

M. Jolivet, O. Dauteuil, A. Dia, Mélanie Davranche, Anne-Catherine Pierson-Wickmann, L. Barrier, M Murray-hudson, Nashaat Mazrui, Remi Marsac, Feng Cheng, et al.

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## REPLY

10.1029/2023GC011315

### Key Points:

- The chemical composition of the alkaline groundwater cannot reflect simple evaporation processes
- The variable origin of islands in the Okavango Delta makes for variable bio-geochemical processes
- The hydro-geochemical functioning of the Delta will only be understood through interdisciplinary studies

### Correspondence to:

M. Jolivet,  
marc.jolivet@univ-rennes1.fr

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### Author Contributions:

**Conceptualization:** M. Jolivet, O. Dauteuil, M. Murray-Hudson  
**Formal analysis:** M. Jolivet, O. Dauteuil, M. Murray-Hudson  
**Writing – original draft:** M. Jolivet, O. Dauteuil, A. Dia, R. Marsac  
**Writing – review & editing:** M. Davranche, A.-C. Pierson-Wickmann, L. Barrier, M. Murray-Hudson, N. Mazrui, Feng Cheng

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## Reply to Comment by Terence McCarthy and Marc Humphries on “Highly Contrasted Geochemical Pattern in Sediments of the Okavango Delta, Botswana Driven by Dust Supply, Hydrological Heritage and Biogeochemical Reactions”

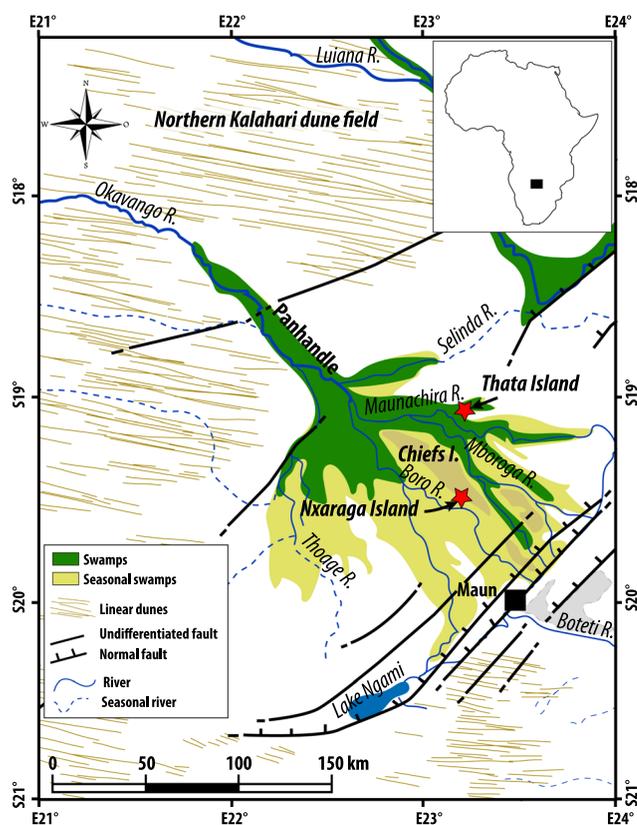
M. Jolivet<sup>1</sup> , O. Dauteuil<sup>1</sup> , A. Dia<sup>1</sup> , M. Davranche<sup>1</sup> , A.-C. Pierson-Wickmann<sup>1</sup>, L. Barrier<sup>2</sup> , M. Murray-Hudson<sup>3</sup> , N. Mazrui<sup>3</sup> , R. Marsac<sup>1</sup>, Feng Cheng<sup>4</sup> , and Xiangzhong Li<sup>5</sup> 

<sup>1</sup>Géosciences Rennes—UMR CNRS6118, University Rennes, Rennes, France, <sup>2</sup>Laboratoire de Tectonique et Mécanique, IPGP, University of Paris, Paris, France, <sup>3</sup>Okavango Research Institute, University of Botswana, Maun, Botswana, <sup>4</sup>Key Laboratory of Orogenic Belts and Crustal Evolution, School of Earth and Space Sciences, Ministry of Education, Peking University, Beijing, China, <sup>5</sup>Key Laboratory of Earth System Science, Yunnan University, Kunming, China

**Abstract** In southern Africa, the Okavango Delta is a large alluvial fan sustaining a unique wetlands ecosystem in the heart of the Kalahari desert. The Delta is fed by an annual flood that brings water from the Angolan plateaus to the north, down to the Okavango region. The fan is characterized by vegetated, permanent or seasonal floodplains, dotted with thousands of sandy islands of various origin (inverted paleo-channels, paleo-dunes, meander bars, etc). On at least some of these islands, shallow groundwater is characterized by a high pH (>8) and a high concentration of metals and metalloids, making it naturally toxic. In the 1990s, McCarthy and co-workers explain the chemistry of that water through an evapotranspiration model, the flood-water being pumped by the vegetation toward the center of the islands and progressively enriched in elements by evapotranspiration. This model has been widely applied to all islands in the Okavango, not considering their individual specificities nor that of the surrounding floodplain. Our work, based on a multidisciplinary approach including sedimentology, mineralogy and geochemistry, and discussed by McCarthy and Humphries, questions the established model suggesting that the alkaline water cannot derive from simple concentration of the floodwater.

## 1. Introduction

Developing in a semi-arid climate and sustaining unique aquatic and terrestrial ecosystems, the Okavango Delta is one of the last pristine sub-tropical wetlands in the world (Figure 1). It has been widely studied, especially since the 1970s, in terms of ecological, geological, hydrological and geochemical processes. Among these investigations, we would like to acknowledge the significant contribution made by the group from the University of the Witwatersrand and associated researchers. This group developed a hydro-geochemical model to explain the fate of the estimated 380,000 tonnes of solutes that enter the Delta each year (McCarthy, 2006) and the occurrence of alkaline water below the central part of the islands (McCarthy, McIver, & Verhagen, 1991; McCarthy et al., 1993). The model proposes that proto-islands in the alluvial fan grow by aggradation from precipitation of mainly calcium and magnesium carbonates and amorphous silica (McCarthy et al., 2012). Plants colonizing the islands are suggested to play a major role in this process by pumping water from the floodplain, enhancing groundwater circulation from the floodplain toward the islands and concentrating minerals in the rim of the islands, leading to their growth. The highly soluble salts, especially sodium bicarbonate, would accumulate below the central part of the islands, forming brines that ultimately sink due to their density contrast with the surrounding water in the aquifer (Bauer et al., 2006; Bauer-Gottwein et al., 2007). The occurrence of such alkaline water below the center of the islands would explain the peculiar vegetation pattern of some of those islands, made of a ring of large trees planted on a ridge ringing the islands, an area covered with small shrubs and finally a barren central zone, sometimes covered with grass. The model is based on sediment coring, major-element and stable-isotope (C and O) analysis of sediment and water, measurement of hydrological parameters (piezometry, water discharge) and geophysical imaging. The results of Jolivet et al. (2023), and previously of Dauteuil et al. (2021), challenge this model with new sedimentological and geochemical data. We suggest that the alkaline water is hosted in a confined aquifer, possibly inherited from a previous phase of construction of the alluvial fan, and capped by a layer of authigenic clay.



**Figure 1.** Map of the Okavango Delta and its surrounding region (modified from Jolivet et al. (2023)).

## 2. Nature of the Islands and Floodplain Dynamics

The exact onset of deposition of the Okavango alluvial fan is still poorly known although some Optically Stimulated Luminescence data suggest that it started around 40 ka. The initiation of the fan followed an episode of flooding, especially marked by over-washing of aeolian dune sand some 100 ka ago (Ringrose et al., 2008). The dune pattern is still preserved in the toe of the fan immediately north of Maun town (Figure 1). In that region, the islands are generally elongated west-east, parallel to the direction of the major dune fields of the northern Kalahari (Thomas, 1988; Thomas et al., 2000). During the last 40 ka, the hydrodynamics of the fan varied several times, following regional climate changes. Tooth et al. (2021) highlighted a period of high discharge in the 7 to 4 ka interval, leading to the formation of >100 m wide meandering channels. Remnants of some of these channels are still preserved in the topography. The present-day channels are a few meters to <20 m wide and generally incise the floodplain topography (McCarthy, Stanistreet, & Cairncross, 1991; McCarthy et al., 1997). Those channels are avulsing, changing their course. This avulsion mechanism is suspected to be largely controlled by vegetation developing on both banks and leading to the formation of levees that ultimately fail, creating a new water course (McCarthy et al., 1992; Stanistreet et al., 1993).

The origin of the thousands of islands that form the Okavango alluvial fan is thus very variable. Some, especially north of Maun town derive from paleo-dunes that have been dissected by the invading channels, others are remnants of meander bars accumulated by large paleo-channels (possibly those described by Tooth et al. (2021) at 7 to 4 ka), some are large portions of land emerging today from the floodplain but supporting evidence of paleo-channels, some may be either paleo sand-bars or inverted channels, and finally a large proportion of them have no clear origin, formed through multiple dissection episodes of surface morphologies. Given their highly

variable origin, each island must have a specific sedimentary structure with varying grain size and sediment facies. Although the previous studies performed by the authors of the comment and their co-workers have investigated several islands (they claim the description of 18 islands through up to 200 auger holes), no detailed description of the sediment has been provided. An exception is formed by indications on the type of material from few islands such as in McCarthy and Ellery (1995). The authors of the comment refer to deep boreholes in Thata Island (Figure 1) but the provided reference is an unpublished PhD thesis from ETH Zurich, for which only a short abstract can be found. This variability in sediment structure of the islands remains to be assessed in detail by precise descriptions of sediment logs. We initiated such a study in the work discussed here (Jolivet et al., 2023), which presents—to our knowledge for the first time—a log description of the boreholes, including grain-size variations, mineralogy and observable sediment structures. The detailed sediment parameters (porosity, mineral composition, amount of organic matter, etc) of the islands certainly plays a major role in the hydro-bio-geochemical processes that control the local composition of the groundwater (e.g., the role of micro-organisms would be very interesting to explore). In that respect, the comment that “None of these studies have revealed a stratigraphic break between an upper sand layer and a lower clay-rich layer” is contradicted by our description of the sediment, the grain size analysis and the  $Al_2O_3$  content, three independent markers that corroborate a stratigraphic change, also marked by the Sr stable isotopes. The authors of the comment suggest that the  $Al_2O_3$  content may be controlled by termite activity. This is indeed a possibility, but aiming at a geochemical analysis we have been very careful to avoid termite mounds, and as stated above, Al is not the only marker we used. We agree with the authors that clay is largely airborne from the Makgadikgadi pan to the SE of the Delta (this we conclude from the Sr isotopes (Jolivet et al., 2023)), but once again, the use of this clay by termites would not lead to the layered stratigraphic pattern we observed, nor to the conjunction of this layer with the water table and the incipient carbonate layer. Finally, the aggradation process (i.e., the growth of the islands through mineral precipitation) proposed by the “brine drop” model (McCarthy et al., 2012), which indeed nicely explains the fate of the solutes entering the Delta will vary from one type of structure to the other, depending on sediment porosity,

permeability and mineralogy. Altogether, the topography of the Delta, can be explained by the variable origin of the islands combined with the dynamics of the rivers, and does not necessarily require aggradation.

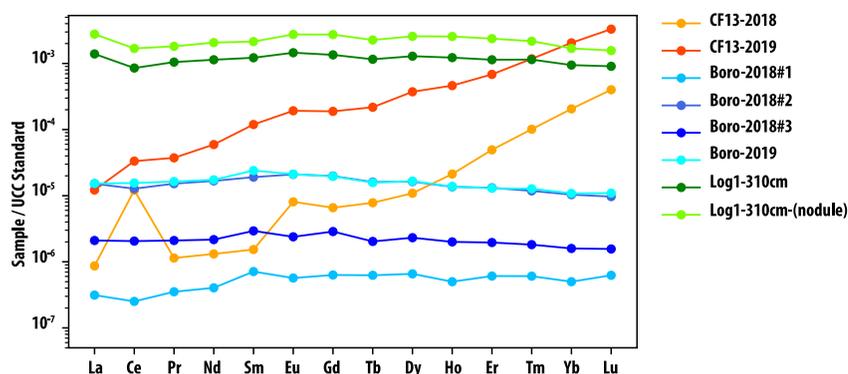
The dynamics of the floodplain is also variable in the Delta: the Panhandle region (Figure 1) and the upper reaches of the alluvial fan are flooded most of the time. The floodplains along the Boro river (western side of the central Chiefs Island) are only flooded during a few months (generally May to October) and dry the rest of the year, some water pools being filled during the rain season or preserved from complete drying. The vegetation composed of grass and reeds dies during the dry season. The Maunachira and Mboroga rivers, east of Chiefs Island, are permanent and a large portion of the floodplain remains flooded, supporting a permanent, thick vegetation of reeds and papyrus (Figure 1). In that respect, Nxaraga island is not situated “*at the lower fringe of the permanent swamp*” as indicated in the comment but indeed in the region of semi-permanent flooding, west of Chiefs Island. This difference in hydrological dynamics has an impact on the concentration of salt, both on the ground and in groundwater (McCarthy & Metcalfe, 1990).

### 3. Salt and Alkaline Waters

Dauteuil et al. (2021) published a first series of stable and trace element geochemical data from water samples from both surface and groundwater in Nxaraga Island, in the SW seasonal swamps of the Delta (Figure 1). They demonstrated, based on the Rare Earth Element (REE) pattern that the alkaline groundwater could not be obtained from concentration through evaporation or evapotranspiration of the surface freshwater. Dauteuil et al. (2021) also showed, based on water-table measurements from October 2017 to January 2019 using a line of 13 piezometers over a horizontal distance of 300 m, that the two aquifers (surface and groundwater) had independent hydrological cycles. In comparison, McCarthy and Ellery (1994) performed water-table measurements over a similar period using 4 non-equipped drilling sites over a distance of 250 m, that they regularly re-opened for measurement. We thus suggested the occurrence of two different, non- or poorly connected aquifers. This conclusion has been re-enforced in our publication (Jolivet et al., 2023) based on a complete analysis of the sediments (stratigraphy, sediment structure, mineralogy, grain size, major and trace elements geochemistry, C, Sr, Nd stable isotopes analysis) hosting the aquifer.

McCarthy and Metcalfe (1990), indicated that “*trona-encrusted islands tend to be more common in the permanent swamps than in the seasonal swamps*” and McCarthy et al. (1986) reported that “[salt] *generally occurs on the fringes of islands as well as on the margins of pans, but occasionally may be found in the interior of smaller islands.*” During two field-expedition in July 2022 and July 2023, we indeed observed frequent salt efflorescence on islands of variable size (from a few 100 m<sup>2</sup> to several km<sup>2</sup>) in the permanent swamps along the Maunachira and Mboroga rivers (Figure 1). This salt is generally found along the shore of shallow swamps, connected to the main rivers by a small (<1 m wide, <50 cm deep) channel. The swamps, and the shores are vegetated, mostly with grass and reeds but also with bushes and sometimes trees. Several authors suggested that these salts, as well as solutes concentrated in the swamps, are washed out of the Delta during the rainy season (November to March) and annual flooding (June to September) when hydraulic connectivity is established between the rivers and the swamps (Letshele et al., 2023; Ramatlapeng et al., 2021, 2023). This may partly explain the fate of the solutes entering the Delta, although we agree that the model should be further developed, especially by establishing the final sink of those solutes. Each year from 2018 to 2023 we visited many islands along the Boro river (i.e., in the seasonal floodplains of the western Delta), at two different periods of the year (June–July and October–December) and could not observe any occurrence of salt on the surface, neither in the central part of the islands nor on the shores like in the east. As reported in Dauteuil et al. (2021) and Jolivet et al. (2023), geochemical analyses of surface samples (down to about 2 m) show low (<0.5% weight) concentrations in Na<sub>2</sub>O or K<sub>2</sub>O that could trace the occurrence of trona, thermonatrite or sylvinitite, the main mineral species observed in the efflorescences (McCarthy et al., 1986). Assuming the occurrence of such salt on the surface, the geochemical pattern reported in our studies would require a perfectly complete leaching of those elements by rain water to a depth of at least 2 m. Our sampling was done in November 2018, before the onset of the rain season, and in July 2019 during the dry season, minimizing the chances of such leaching.

Finally, we do not fully understand the comment about partitioning of REEs into calcite, as this is exactly what we propose: “*The REE patterns in the sediments are controlled by carbonate nodules (Figures 7 and 10)*” (see Section 5.1.2 of Jolivet et al., 2023). We also indicate that, at high pH and high carbonate concentration, organic matter most probably plays a role in shaping the REE pattern, especially by favoring the partitioning of Ce toward



**Figure 2.** Rare earth element spectrum (normalized to UCC standard) for water and sediment from Nxaraga Island and the Boro river. CF13-2018 (Dauteuil et al., 2021) and CF13-2019 (this study, Table 1) are groundwater samples from the center of the island collected in November 2018 and July 2019; Boro-2018#1, #2, and #3 are water samples from the Boro river or related channels taken in November 2018 (see samples OkaRiv, ChaNx and ChaAv in Dauteuil et al. (2021)); Boro-2019 is a water sample from the Boro river taken in July 2019 (this study, Table 1); Log1-310 cm is a sediment sampled at 310 cm depth in the center of Nxaraga Island (in the water table compartment) and Log1-310 cm-(nodule) is a carbonate nodule sampled at the same location (see data in Jolivet et al. (2023)).

the water (through adsorption on colloids), leading to the negative Ce anomaly in our soil samples. The calculation of partition coefficients ( $D^{REE}$ ) proposed in Zhong and Mucci (1995) and Voigt et al. (2017) are of great interest but the experiments were conducted at neutral pH instead of the high pH values (>8) observed in Nxaraga with incipient calcrete developing. The pH plays a role in modifying the solid-liquid  $D^{REE}$  (Dia et al., 2000; Sholkovitz, 1995), especially in the carbonate system, in which a competition between REE incorporation/adsorption into the solid phase and REE complexation by dissolved carbonates occurs. Sawada et al. (2005) investigated the effect of pH and observed an increase of  $D^{Gd}/D^{Yb}$  with increasing pH, which should be related to the solution speciation of REE. With increasing pH,  $REEHCO_3^{2+}$  that weakly fractionate REE because complexation constants for all REE are similar, will give way to  $REE(CO_3)_2^-$  whose stability constant drastically increases with atomic number (Luo & Byrne, 2004), hence favoring the occurrence of HREE in the aqueous phase (Pourret et al., 2007). It has also been shown that dissolved organic matter induces similar effects as carbonates (more pronounced HREE complexation at high than at low pH), which can be explained by the joint effects of site heterogeneity and cation competition (Marsac et al., 2010, 2011, 2012, 2013). As indicated in Jolivet et al. (2023), in the water table compartment, “[...] water and sediment are at equilibrium: the groundwater REE pattern exhibits both a strong HREE enrichment and a positive Ce anomaly.” Figure 2 shows the REE patterns of water samples from the Boro river, from the groundwater in the center of Nxaraga Island and of two representative soil and carbonate nodule samples from the water table compartment (see Table 1, Dauteuil et al., 2021; Jolivet et al., 2023 for data). The Boro river water, the sediment and carbonate nodules are HREE depleted with  $2.18 < Gd/Yb < 3.31$  and  $2.47 < Gd/Yb < 2.80$  values respectively. Comparatively, the groundwater is enriched in HREE with  $0.06 < Gd/Yb < 0.16$  values. These data show that the REE pattern of the groundwater is largely controlled by solute carbonates and most probably organic matter, that specifically complex HREE.

#### 4. Conclusion

Our definition of two different hydrological compartments is not solely based on REE patterns, but on the combination of all the independent data acquired (clay content,  $^{87}Sr/^{86}Sr$  ratio (indicating massive dust input from the

**Table 1**  
Rare Earth Elements Concentrations in ppm for the Groundwater Collected in the Central Part of Nxaraga Island in July 2019 and the Boro River Water Collected in July 2019

Sample	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
CF13-2019	0.366	2.133	0.265	1.543	0.537	0.169	0.171	0.139	1.306	0.370	1.572	0.388	4.512	1.059
Boro-2019	0.459	0.995	0.116	0.452	0.108	0.019	0.075	0.010	0.058	0.011	0.030	0.004	0.024	0.004

Makgadikgadi pan), major and trace element concentrations,  $\delta^{13}\text{C}$  ratio on water and soil organic matter, hydrology). We do appreciate the significance of the work already done on the Delta but we are of the opinion that our new data should be the base to question at least the unicity of the “brine drop” model. As explained above, the sedimentology and hydrology of the Delta is complex and variable from one location to the other. The nature of the islands, their sedimentological structure and composition, as well as the hydrology and ecology of the surrounding alluvial plains, should generate differences in hydro-geochemical functioning. Finally we agree with the authors of the comment that “[...] *further understanding will only be obtained by large-scale interdisciplinary research.*”; an approach we are now taking.

### Data Availability Statement

The geochemistry used in this reply are all available in Table 1 or in published works (Dauteuil et al., 2021; Jolivet et al., 2023).

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