Enhanced silicate chemical weathering of the South African Plateau and its contribution to the global climate cooling trend during the late Cretaceous

Camilo E. Gaitan[1,2], Emmanuelle Pucéat[2*], Pierre Pellenard[2], Emmanuelle Venin[2], Fabrice Monna[3], Germain Bayon[4], Thierry Adatte[5], Cécile Robin[6], and François Guillocheau[6]

[1]*Climerock, Gif-sur-Yvette, France*

[2]*Biogéosciences Dijon, Université Bourgogne Franche – Comté, UMR CNRS 6282, Dijon, France.*

[3]*ARTEHIS, UMR, CNRS, 6298, Université de Bourgogne–Franche Comté, Dijon, France*

[4]*Unité de Recherche Géosciences Marines, IFREMER, Plouzané, France.*

[5]*ISTE, Université de Lausanne 1, Lausanne, Switzerland.*

[6]*Géosciences Rennes, Université de Rennes 1, UMR CNRS 6118, Rennes, France.*

*Corresponding author: emmanuelle.puceat@u-bourgogne.fr

Abstract

Climate cooling during the late Cretaceous (ca. 100-66 Ma) has been linked to CO_2 withdrawal due to enhanced denudation episodes recorded in the East-South American and West-South African margins. However, the onset of these episodes is recorded after the main cooling stage (i.e., Turonian-Santonian — ca. 93-83 Ma). Here, we investigate the clay fraction (< 2 μm) of sediments from the borehole O-A1 located in the continental slope of the Cape Basin, adjacent to the South African Margin. We combine these results with previous records from the basin (DSDP 361) to investigate the controlling factors in the denudation record along the South

African Plateau and their relationship with the climate cooling during the late Cretaceous. The $\Delta\varepsilon_{Hf}$ (proxy for silicate weathering intensity) record from site O-A1 shows an increase in silicate chemical weathering during the Cenomanian-Maastrichtian (ca. 95-68 Ma) interval, predating the Campanian-Danian interval shown by the DSDP 361 record, which offset to the site O-A1 is primarily linked to age model uncertainties from DSDP 361. This increase in silicate chemical weathering is concomitant to an enhanced phase of physical erosion and tectonic uplift of the South African Plateau as well as global climate cooling, suggesting this enhanced silicate chemical weathering phase is tectonically driven. Our $\Delta\varepsilon_{Hf(80)}$ data from site O-A1 shows for the first time an increase in silicate chemical weathering as early as the Cenomanian, suggesting not only that it could have played a role maintaining the cool global climate trend during the late Cretaceous, but that this episode could have contributed to trigger the CO_2 withdrawn responsible for this cooling that started by the Cenomanian-Turonian. Additionally, we suggest that this enhanced silicate chemical weathering episode along the South African Plateau might have influenced atmospheric CO_2 concentrations and climate through two different mechanisms, initially via silicate reactions, but also through increased nutrient riverine input into the basin, which could have contributed to enhanced primary productivity and organic carbon storage as shown by the increase in Ba_{excess} during the Santonian-Maastrichtian (ca. 85-65 Ma).

Keywords: Isotope geochemistry, Tectonics-Climate, Silicate weathering, South African Plateau, Cretaceous cooling, Marine productivity.

1. Introduction

Sedimentary basins represent the best archive to track the long-term evolution of denudation processes in ancient environments. Recent studies conducted in basins adjacent to the East-South American and West-South African margins relate tectonic activity with enhanced denudation episodes, subsequent CO_2 withdrawal, and climate cooling (Corentin et al., 2024, 2023, 2022; Gaitan et al., 2023). These studies have shown through clay mineralogy and isotopic records (Hf-Nd isotopes on clays) increases in silicate chemical weathering during the late Cretaceous, which is recognised as one of the most important process for CO_2 withdrawal (Berner et al., 1983; Gaillardet et al., 1999; Hilton and West, 2020). While these weathering episodes have been associated with the long-term climate cooling trend observed in the late Cretaceous (Friedrich et al., 2012; O'Brien et al., 2017), Hf-Nd isotopic records reveal increases between the Santonian-Campanian interval in the South American margin (Corentin et al., 2023, 2022), and between the Santonian-Maastrichtian interval in the West-South African margin (Corentin et al., 2024; Gaitan et al., 2023), both later to the suggested onset of the cooling trend around the Turonian (Friedrich et al., 2012; O'Brien et al., 2017). In the context of the South African Margin, the proposed increase in chemical weathering relies only on the geochemical record from DSDP core 361. Yet this site presents relatively large uncertainties in the age model of the studied sediments (Gaitan et al., 2023), hindering the identification of a precise timing for the onset of the enhanced weathering episode. Additionally, no major changes in chemical weathering along the South African margin are detected at DSDP core 361 during the Turonian-Campanian interval, which is the main cooling phase reported by global isotopic records during the late Cretaceous ($\delta^{18}O$ benthic) (Friedrich et al., 2012; O'Brien et al., 2017). Therefore, although the recorded weathering episodes along the East-South American and West-South African margins could have contributed to the

cooling conditions during the late Cretaceous, the mechanisms responsible for the cooling onset remain elusive.

The evolution of the chemical weathering signal from DSDP core 361 relies on the analysis of detrital clay minerals coming predominantly from the South African Plateau and depositing in the abyssal plain of the Cape Basin (Gaitan et al., 2023). However, due to the distal location of the sedimentary record, the incorporation of allogenic sediments by deep-bottom currents is not entirely discarded (Murphy and Thomas, 2013; Natland, 1978), and differential settling processes on clay minerals might also be involved introducing some bias into the paleoclimatic interpretation (Gibbs, 1977; Petschick et al., 1996). Multiple thermochronological data — Apatite-Fission Track (AFT) and Apatite (U-Th)/He (AHe) — have shown that regional structures across the Plateau are important in the rock cooling events during the late Cretaceous (Green et al., 2017; Kounov et al., 2009, 2013; Raab et al., 2002; Stanley and Flowers, 2020; Tinker et al., 2008; Wildman et al., 2015, 2021). These regional structures could have modified the source areas of the denudation episodes (i.e., erosion and weathering) inferred from these rock cooling events. These denudation episodes have been further linked to tectonic uplift involving continental tilting (Baby et al., 2018a, 2019; Braun et al., 2014; Stanley et al., 2021), responsible for drainage reorganization (De Wit et al., 2009; De Wit, 1999; Goudie, 2005; Stevenson and Mcmillan, 2004), and by extension for variations in the sediments source areas. Estimations of sediment fluxes and volumes have also shown that the tilting component of the uplift appears to have affected the sediment distribution along the basins located adjacent to the South African margin (Baby et al., 2019). Altogether, these processes affecting the source and distribution of the sediments deposited in the Cape Basin could have impacted the denudation record, particularly the chemical weathering, and hinder any interpretation of the impact of these processes in the climate during the late Cretaceous. For this reason, a process-response comparison of the denudation record along the Cape Basin appears of prime

95 importance to understand the dynamics controlling this record and the implications for climatic
96 interpretations.
97 The aim of this study is to investigate the sedimentary record at the continental slope of the
98 Cape Basin (borehole O-A1— Fig. 1) and compare it with previous records from the abyssal
99 plain (DSDP core 361— Fig. 1; Gaitan et al., 2023). Through this comparison we study the
100 impacts of changes in sediment distribution and sources in the erosional and chemical
101 weathering signal recorded along the Cape Basin, also exploring the links of this record to the
102 global climate cooling during the late Cretaceous. To do so, we combine X-ray diffraction
103 (XRD) analyses, major and trace elements, Transmission Electron Microscopy (TEM), and Hf
104 – Nd isotopic compositions on clay fractions (< 2 μm - Bayon et al., 2016). Through this suite
105 of techniques, we seek to understand the dynamics affecting geochemical signals, specifically
106 on the clay fraction across the sediment routing system.
107

108 ## 2. Geological Setting

109 ### 2.1 Evolution of the southwestern African margin
110

111 The South African margin evolution began with the rifting between the South American
112 and African Plates and the subsequent opening of the Atlantic Ocean during the late Jurassic –
113 early Cretaceous (Moulin et al., 2010; Torsvik et al., 2009). It has been suggested that this
114 opening started at the southern segment and moved northwards (Koopmann et al., 2016;
115 Rabinowitz and LaBrecque, 1979), but the rifting onset in the southern segment is not yet fully
116 understood due to age uncertainties on the magnetic anomalies (e.g., Baby et al., 2018b; Moulin
117 et al., 2010). However, it is acknowledged that magnetic anomaly M3 at 130 Ma marks the
118 rifting-drifting transition in this segment (Collier et al., 2017). Deposition during this period is

marked by large volcanic activity linked to the Paraná-Etendeka Large Igneous Province Province (ca. 135-133 Ma; Gomes and Vasconcelos, 2021; Renne, 2015), this is supported by the presence of Seaward-Dipping Reflectors (SDRs) overlain graben and half graben basement structures (Gladczenko et al., 1997). Regional Valanginian-Hauterivian fault-bounded syn-rift sequences are present across the southwestern African basins (Séranne and Anka, 2005). These sequences comprise lava flows with continental sediments, essentially sandstones and claystones (Hirsch et al., 2010; Jungslager, 1999; Van Der Spuy, 2003).

Post-rift deposits consist of thick siliciclastic sequences of about 3 to 5 km distributed along the 4 basins adjacent to the margin: Walvis, Lüderitz, Orange, and Cape (Aizawa et al., 2000). (Baby et al., 2018b) defined three main first-order depositional phases for these post-rift deposits. The first phase is a retrogradational stage (131-93.5 Ma) marked by an increase rate of accomodation space compared to sediment supply. The creation of this accomodation space is associated to the thermal subsidence of the margin. During the second stage (93.5-66 Ma), deposition shifts to a predominant progradational-aggradational phase, whereas the margin is tilted seawards (Aizawa et al., 2000; Baby et al., 2019). A pronounced increase in sediment supply related to the tectonic uplift of the South African Plateau is linked to this depostional change. The final stage (66-0 Ma) represents primarily a retrogradational phase, although there is a progradational stage within this period at ca. 27 Ma (Baby et al., 2018b). The retrogradational stage is likely associated with the stabilization of the inland Plateau and climate aridification, which led to a decrease in sediment supply.

The stratigraphy of the margin is divided in 8 units bounded by stratigraphic markers (e.g., unconformities or maximum flooding surfaces) that define relevant regional deposition cycles (Baby et al., 2019, 2018b). Lithology of this post-rifting units comprises mainly siltstones, although limestones are also an important component in the Unit 1 (130-113 Ma) from the Walvis and Lüderitz basins (Baby et al., 2019).Cape Basin

2.2 Cape Basin

The Cape Basin is divided in 8 units and bounded by the regional stratigraphic surfaces defined in the basins along the margin. The Cretaceous interval in the basin spans between Unit 1 and 5 (Baby et al., 2018b). Unit 1 is bounded by the unconformities 6At1 (130 Ma) and 13At1 (113 Ma), the former representing the breakup unconformity whereas the latter a depositional hiatus (Paton et al., 2008, 2007). This unit comprises a sedimentary package evidencing the rifting-drifting transition of the margin. At the base, aeolian sandstones with basalts are covered by carbonate cemented marine sandstones. On top, shelly sandstones with some local lagoonal limestones are topped by marine shales (Baby et al., 2018b). The upper boundary of Unit 2 is the seismic marker 14At1 (100 Ma). This unit is dominated by silty-claystones overlain by shales that define marker 14At1 (Baby et al., 2018b). This unit comprises a predominant retrogradational trend (Baby et al., 2019), but most importantly evidences sediment supply from two disconnected deltaic systems located southern of the modern Orange river system (Baby et al., 2018b). Unit 3 consists wholly of deep marine shales (Baby et al., 2018b; Paton et al., 2008), with marker 15At1 (93.5 Ma) as the upper boundary. Depositional trend in this unit still is mostly retrogradational, marked by wedges backstepping (Baby et al., 2019). The upper boundary of Unit 4 is marked by the unconformity 17At1 (81 Ma), which has been interpreted as a major sea-level fall. Claystones dominate in this unit in a predominantly aggradational depositional trend. The top of Unit 5 is marker 22At1 at the Maastrichtian-Paleogene boundary (66 Ma). Depositional trend changes from aggradational to progradational, linked to variations in the ratio of subsidence and sediment supply (Baby et al., 2018b; de Vera et al., 2010). Lithology at this unit is dominated by clayey siltstones (Baby et al., 2018b). Unit 6 lies on top of the Cretaceous sequence and it is bounded on top by an Oligocene unconformity (Baby et al., 2018b; de Vera et al., 2010). This unit displays deltaic progradational units (Baby et al., 2018b; de Vera et al., 2010).

3. Material and methods

3.1 Borehole description

In this study we analysed 91 cutting samples from the borehole O-A1 provided by the South African Petroleum Agency. This borehole is located at the continental slope of the Cape Basin (Fig. 1; 33.161539°S 16.822406°E), around 100 km off the southwestern African margin. The borehole comprises a stratigraphic succession of about 3650 m, spanning from the Aptian - Barremian to the middle Eocene period (Valicenti et al., 1993), but the samples analysed in this study span between the Albian and Ypresian periods (i.e., 115-55 Ma), representing Unit 1 to 5 from those described by Baby et al. (2019) (Fig. 2). The basement of the borehole consists of volcanic rocks presumably Hauterivian in age (ca. 132 Ma; Valicenti et al., 1993). On top, there is an Aptian-Albian sequence of argillaceous sandstones (ca. 125-100 Ma), with increased clay abundance at the top and the base. At the late-early Cretaceous boundary (100 Ma), the predominant lithology are claystones interbedded with sandstone layers (Dudus et al., 1992), some minor dolomite stringers are also present in this interval. Throughout the late Cretaceous (100-66 Ma) the succession consists of claystones with occasional pyrite, and stringers of limestones, dolomites, and siltstones (Dudus et al., 1992). Siltstones stringers are predominantly argillaceous and glauconitic. The interval between the Paleocene and the middle Eocene (ca. 66-50 Ma) consists of claystones with sporadic fossiliferous limestones and siltstone stringers (Dudus et al., 1992). At the base of this interval, stringers are scarce and claystones are more calcareous, dolomite stringers are present as well. Sporadic pyrite and glauconite occur within the claystones along the whole interval (Dudus et al., 1992).

192 The selected samples were targeted considering even distribution within the studied time
193 interval (i.e., 115-55 Ma) and clay content. To ensure an even distribution of the analysed
194 samples an age model (Supplementary material FS1) was built based on the planktonic
195 foraminifera report by Valicenti et al. (1993).

196 There were no elements to determine neither the Aptian-Albian boundary (113 Ma) or the
197 Albian stage (113-100.5 Ma), which could be missing. Only the occurrence of *Globuligerina*
198 *hoterivica* around 4170 mbsf indicates late Aptian ages, so the Aptian-Albian boundary was
199 tentatively placed at this depth (113 Ma). A late Albian-Mid Cenomanian (ca. 105-97 Ma)
200 interval was defined around 3624 mbfs based on the last occurrence of *Rotalipora appenninica*.
201 The Cenomanian-Turonian boundary (93.9 Ma) was placed at 3570 mbsf based on the
202 appearance of *Helvetoglobotruncana helvetica*. The last occurrence of this assemblage
203 indicated a Mid-Turonian age (ca. 91 Ma) at 3408 mbsf, but there were no elements to define
204 the Turonian-Coniancian boundary (89.8 Ma). An increase in the occurrence of
205 *Marginotruncana coronata* and the appeareance of *Globotruncana* species enabled to place an
206 interval Coniacian-Santonian (89.8-83.6 Ma) around 2706-2751 mbsf, still the boundary
207 cannot be determined with certainty. Following the occurrence of *Globotruncanita elevata,*
208 distinctive of Campanian age, the Santonian-Campanian boundary (83.6 Ma) was placed at
209 2697 mbsf. The occurrence of *Globotruncanita aegyptiaca* indicative of a late Campanian age
210 (ca. 75 Ma) and the last occurrence of *Globotruncana ventricosa* enabled to place the
211 Campanian-Maastrichtian boundary (72.1 Ma) around 2580 mbfs. The presence of
212 *Abathomphalus mayaroensis* around 2553 mbsf is indicative of a late Maastrichtian age. No
213 characteristic Danian fauna was identified. The *Globanomalina chapmani* characteristic of a
214 Selandian-Thanetian age was observed around the interval 1550-1340 mbsf, however it was
215 joint by species like *Globigerinatheka index,* which is characteristic of Eocene (ca. 40 Ma).

The boundary Maastrichtian-Danian (66 Ma) was then placed at 2553 mbsf, and the subsequent Cenozoic upper interval is tied to an Eocene age (33.9 Ma) around 1340 mbsf.

Based on these planktonic foraminifera fossil assemblages, intervals and age boundaries were established following the ICS chronostratigraphic chart v2023/06 (Cohen et al., 2013; Updated 2023). The age model was built using linear regressions between seven tie-points: the Aptian-Albian boundary (113 Ma; 4170 mbsf), the Mid-Cenomanian (97.2 Ma; 3624 mbsf), the Cenomanian-Turonian boundary (93.9 Ma; 3570 mbsf), the Santonian-Campanian boundary (83.6 Ma; 2697 mbsf), the Campanian-Maastrichtian boundary (72.1 Ma; 2580 mbsf), The Maastrichtian-Danian (66 Ma; 2553), and the Eocene stage tentatively at 1340 mbsf (ca. 34 Ma).

3.2 Analytical techniques

3.2.1 *X-ray diffraction (XRD) analyses*

XRD analyses were performed for 77 samples for both bulk rock and clay fractions (< 2µm) in the Biogéosciences Laboratory (UMR CNRS 6282) at the University of Burgundy, France (Supplementary table S1). The bulk rock samples were grinded and set in disks adapters whereas the clay fractions were prepared as oriented glass slides. Both set of measurements were performed on a Bruker D8 Endeavor diffractometer with Cu-Kα radiations, a LynxEye detector coupled to a Ni filter, and under 40-kV voltage with 25-mA intensity. For the clay fraction (< 2µm), each oriented glass slide was measured three times following the protocol set by Moore and Reynolds (1989), including both ethylene glycol and heating (490 °C) preparation stages. The main intensity peaks were identified using the MacDiff software 4.2.5 (Petschick, 2001), and the mineral proportions, both for bulk and clay fractions, were determined by calculating the area under the basal reflections peaks (d_{001}). For the clay

fractions (< 2µm) this calculation was done using the resulting diffractograms from the ethylene glycol treatment (Moore and Reynolds, 1989).

3.2.2 *Transmission Electron Microscopy (TEM) observations*

TEM analyses were performed on clay fractions from 6 samples (Supplementary table S1) at the Agroécologie Institut from the Institut National de Recherche pour l'agriculture, l'alimentation et l'environnement (INRAE) in the University of Burgundy, France. The samples were dispersed in distilled water and 2% v/v butylamine solution and then set on a carbon formvar Cu grid. Images were taken using a MET Hitachi 4800.

3.2.3 *X-ray fluorescence (XRF) analyses*

Forty samples were processed for major and minor oxides abundances (Supplementary table S1) using XRF at the Institut Sciences de la Terre, at the University of Lausanne, Switzerland. Fused disks were prepared from 1.2 g calcined sample powder mixed with Lithium-Tetraborate (1:5 proportions). The measurements were done using a wavelength dispersive PANalytical Axios^{mAX} spectrometer fitted with a 4 kW Rh X-ray tube. Rock silicate reference materials were used for calibration.

3.2.4 *Chemical processing for trace element and isotopic analyses*

Twenty four samples were grinded and processed (1.3 to 1.5 g of powder) through sequential leaching following a procedure based on the protocols described in Bayon et al., 2002 and Gutjahr et al., 2007 (Supplementary table S1). Carbonates were first removed using 10% v/v acetic acid reacting overnight with the samples. After a rinsing stage with ultrapure 18.2 MΩ water, the Fe-Mn oxyhydroxide phases were removed using a 0.5 M hydroxylamine hydrochloride solution in 20% v/v acetic acid reacting with the samples for 48 hours. After another rinsing stage with ultrapure 18.2 MΩ water, organic matter was removed using a 5%

266 H_2O_2 solution reacting with the samples for 48 hours, to finally be rinsed again with ultrapure
267 18.2 MΩ water. Clay fractions (< 2μm) were separated after this leaching sequence using
268 particle decantation according to the Stokes law.

269 After leaching and sampling of the clay fraction (< 2μm) the samples were dried and
270 processed by alkaline fusion following the protocol described in Bayon et al., 2009. Three 3
271 certified reference standards (BHVO-2, MAG-1, and BCR-2) from the United States
272 Geological Survey (USGS) were also included during the alkaline fusion. Around 50 mg of
273 each sample were placed into carbon crucibles together with 0.6 g of NaOH, 1.2 g of Na_2O_2,
274 and 0.5 g of Tm spike. The crucibles underwent a heating stage at 650 °C during 12 minutes in
275 a furnace, to then be cooled down with ultrapure 18.2 MΩ water, resulting in the precipitation
276 of Fe-hydroxides. The samples were then centrifuged, and finally dissolved in 3,3 ml of 6 M
277 HCl. From this solution, 0.3 ml were extracted for trace elements analyses. The Tm spike was
278 added as an internal standard in the quantification of trace elements following the method
279 described in Barrat et al., 1996.

280

281 3.2.4.1 Trace elements

282

283 Trace elements concentrations were measured by inductively coupled plasma mass
284 spectrometry (ICP-MS) on a Thermo Scientific X-Series II®, at the Pole Spectrométrie Océan
285 in Brest, France (Supplementary table S2). We used standard bracketing with the Tm spike to
286 quantify trace elements as described in Barrat et al., 1996 and Freslon et al., 2011. Accuracy
287 control and reproducibility were assessed analysing the 3 USGS reference standards: BHVO-
288 2, MAG-1, and BCR-2 (Supplementary table S3). Deviations of trace elements from the
289 reference BHVO-2 (Jochum et al., 2016) were below 14%. For MAG-1 and BCR-2 standards,

some elements presented higher deviations, in particular the light rare earth elements (LREE) at around 20% (Supplementary table S3).

3.2.4.2 Hf-Nd Isotopic analyses

Extraction of Hf and Nd for 24 samples was performed by ion chromatography using singled customed columns with AG50W-X8 (200-400 mesh) resin for REE + Hf, Ln Spec resin (50-100 μm) for the Nd fraction, and Ln Spec resin (100-150 μm) for the Hf fraction. Column chromatography was performed applying the adapted separation protocol for Hf and Nd described in Gaitan et al. (2023) for the low pressure, automated column chromatography, PrepFAST-MC® system device. Isotopic measurements were performed using a MC-ICP-MS Neptune Plus® (ThermoFisher-Scientific) at the École Normale Supérieure (ENS) of Lyon, France. Errors are reported as 2 standard deviation (2 sd).

Hafnium isotope measurements performed at the ENS consisted of a static acquisition method of 40 cycles, with an average intensity of 4.3 V throughout the session. Ratios were corrected for mass bias through exponential law and using the stable isotope ratio $^{176}Hf/^{177}Hf$ = 0.7235. Mass bias corrected $^{176}Hf/^{177}Hf$ ratios were normalized to a JMC 475 value of 0.282163 (Blichert-Toft et al., 1997). Repeated measurements of bracketed JMC 475 standards during the analytical session gave a ratio of $^{176}Hf/^{177}Hf$ = 0.282270 ±0.000006 (21 ppm, n = 17). Reference standards BHVO-2 and BCR-2 gave $^{176}Hf/^{177}Hf$ normalised isotopic values of 0.283103 ±0.000020 (70 ppm, n = 3) and 0.282869 ±0.000004 (14 ppm, n = 1) respectively. Both standards within the range of reference values determined by Weis et al. (2005). Four procedural blanks for Hf were also measured, obtaining values between 27 and 44 pg (Supplementary table S3).

Neodymium isotope measurements performed at the ENS consisted of a static acquisition method of 40 cycles, with an average intensity of 5 V. Ratios were corrected for mass bias through exponential law and using the stable isotope ratio $^{143}Nd/^{144}Nd = 0.7219$. Mass bias corrected $^{143}Nd/^{144}Nd$ ratios were normalized to a JNdi-1 value of 0.512115 (Tanaka et al., 2000). Repeated measurements of bracketed JNdi-1 standards during the analytical session gave a ratio $^{143}Nd/^{144}Nd = 0.511980 \pm 0.000009$ (18 ppm, n = 16). Reference standards BHVO-2 and BCR-2 gave $^{143}Nd/^{144}Nd$ normalised isotopic values of 0.512984 ±0.000013 (25 ppm, n = 3) and 0.512644 ±0.000009 (17 ppm, n = 2) respectively. Both standards are consistent with the reference values determined by Weis et al., (2005). Four procedural blanks for Nd were measured as well, obtaining values between 18 and 313 pg, which are approximately $1x10^3$ times lower than the average concentrations obtained for the samples (Supplementary table S3).

Results for hafnium and neodymium isotopic compositions are reported in epsilon notation relative to the deviation from the chondritic reference material (CHUR) reported in Bouvier et al. 2008, $^{176}Hf/^{177}Hf_{CHUR} = 0.282785$ and $^{143}Nd/^{144}Nd_{CHUR} = 0.512630$:

$$\varepsilon_{Hf} = \left(\left[\frac{\left(\frac{176Hf}{177Hf}\right)_{Sample}}{\left(\frac{176Hf}{177Hf}\right)_{CHUR}} - 1\right] * 10^4\right) \text{ and } \varepsilon_{Nd} = \left(\left[\frac{\left(\frac{143Nd}{144Nd}\right)_{Sample}}{\left(\frac{143Nd}{144Nd}\right)_{CHUR}} - 1\right] * 10^4\right)$$

ε_{Nd} and ε_{Hf} values are also reported age corrected (t=80 Ma; Table 1). The ε_{Hf} values were corrected for radioactive decay of ^{176}Lu to ^{177}Hf using the measured Lu and Hf concentrations ($^{176}Lu/^{177}Hf = Lu/Hf * 0.1419$), the ^{176}Lu radioactive decay constant λ ($1.867 \pm 0.008 \times 10^{-11}$ $year^{-1}$; Söderlund et al., 2004), and the present-day $^{176}Lu/^{177}Hf_{(CHUR)}$ value of 0.336 (Bouvier et al., 2008). The ε_{Nd} values were corrected for radioactive decay of ^{147}Sm to ^{144}Nd using the measured Sm and Nd concentrations ($^{147}Sm/^{144}Nd = Sm/Nd * 0.6049$), the ^{147}Sm radioactive decay constant λ (6.54×10^{-12} $year^{-1}$; Lugmair and Marti, 1977), and the present-day

$^{147}Sm/^{146}Nd_{(CHUR)}$ value of 0.1960 (Bouvier et al., 2008). Age corrections were done to 80 Ma to interpret the data. This age represents the average age of the analysed time interval.

ε_{Nd} and ε_{Hf} values from clay fractions display a correlation that is denominated as the "Clay Array" (Bayon et al., 2016). Offsets in ε_{Hf} values from this array are expressed as $\Delta\varepsilon_{Hf}$ (Equation 1) and account for the intensity in chemical weathering (Bayon et al., 2016). $\Delta\varepsilon_{Hf}$ values were calculated using ε_{Nd} and ε_{Hf} values at present time (t = 0) and age-corrected to 80 Ma, these values are reported in Table 1.

$$\Delta\varepsilon_{Hf(t)} = \varepsilon_{Hf(t)} - (0.78 * \varepsilon_{Nd(t)} + 5.23) \quad \text{(Equation 1)}$$

4. Results

4.1 Bulk rock composition and clay mineral assemblages – XRD analyses

Bulk rock composition measured shows a predominance of quartz and phyllosilicates (i.e., clays and micas) along the section, with calcite only becoming predominant after the Santonian (~2800 mbsf; ca. 84 Ma) (Supplementary table S4; Fig. 2). At the base, during the Aptian-Albian interval (ca. 114-110 Ma), quartz content increases reaching values around 88%. Above, quartz content decreases while phyllosilicates content increases, reaching values up to 35% in the Turonian (ca. 90 Ma). An increase in calcite concentrations is observed at the Cenomanian (ca. 96 Ma). Since the Coniacian, calcite content increases in parallel with to a decrease in quartz content, while phyllosilicates proportions remain stable at values around 35%. The increase in calcite content continues up to the Maastrichtian-Paleogene transition, where it reaches values around 50%. Between the Danian and the Ypresian, quartz and phyllosilicates content increase up to values around 35% and 40%, respectively. Calcite content

362 decreases from the Danian onwards reaching to values close to 20%, only showing a positive
363 excursion around the Danian-Selandian boundary with values close to 55%. Plagioclase and
364 K-Feldspar are present throughout the section but in minor proportions. The percentages along
365 the section are around 8%, except for sample O-A1/-91 at the base of the section with 16% and
366 19% for K-Feldspar and plagioclase, respectively. Other minerals like pyrite, halite, and
367 gypsum, are also present along the section but in proportions lower than 10% (Supplementary
368 table S4).
369
370 The clay assemblages at the base of the section are dominated by smectite with values close
371 to 75% (Supplementary table S4; Fig. 2). However, in the late Aptian (ca. 113 Ma), smectite
372 disappears simultaneously to the appearance of illite-smectite mixed layer, which reaches
373 values around 10%. During the Aptian-Albian interval, illite and chlorite increase up to values
374 around 50% and 30%, respectively. By the early Cenomanian (ca. 100 Ma), illite values
375 decrease to around 15%, following an increase in illite-smectite and kaolinite. From the
376 Cenomanian to the Turonian, most of the clay assemblages remain stable, with values around
377 35% for both chlorite and illite, 15% for kaolinite, and around 10% for illite-smectite. Chlorite
378 displays an increase around the Turonian-Coniacian transition and chlorite-smectite mixed
379 layers start to appear in the late Coniacian (ca. 86 Ma), whereas illite decreases to values around
380 20%. By late Coniacian (ca. 87 Ma), illite-smectite and kaolinite disappear concomitantly to
381 the appearance of smectite, with illite also showing a decrease down to 10%. Since, smectite
382 increases steadily up to the Danian (ca. 65 Ma) where it peaks at values around 75%.
383 Concomitant to such increase, chlorite-smectite abundance decreases and reaches values
384 around 25% in the Maastrichtian-Danian transition. Illite oscillates during this period but
385 displays a subtle increase reaching values close to 30% in the Maastrichtian-Danian transition.
386 During the Danian-Ypresian interval, smectite remains the most abundant assemblage with

values close to 70%, while illite and chlorite-smectite decrease significantly to values around 8%, however during the Thanetian, illite showed a continuous increase up to values around 20%.

4.2 Hf - Nd isotopic compositions

The measured ε_{Hf} values range between -4.8 and -2.1 ε-units throughout the section (Fig. 2; Table 1). During the Albian, ε_{Hf} values remain stable around -3 ε-units, only to decrease down to -4.3 ε-units in the Cenomanian (ca. 95 Ma). Above, values show a slight increasing trend up to the late Santonian (ca. 85 Ma), where they reach values around -2 ε-units. During the early Campanian there is a drop in the values down to -3.7 ε-units, follow by a very minor increasing trend that continues up to the early Danian (ca. 65 Ma), where they reach back to values around -2.5 ε-units. Lastly, the values decrease during the Danian-Ypresian to reach a minimum of -4.9 ε-units. Measured ε_{Nd} values range between -9.7 and -8.1 ε-units throughout the section (Fig. 2; Table 1). From the Albian up to the early-Coniacian (ca. 89

402 Ma), ε_{Nd} values display a slight increase from -8.8 to -8.4 ε-units, apart from a negative

403 excursion (-9.7 ε-units) in the late Albian (ca. 102 Ma). Above the Coniacian, the ε_{Nd} values

404 display a long subtle negative trend up to the Thanetian (ca. 58 Ma), where they reach values

405 around -9.7 ε-units. However, in this interval there is a minor increasing period during the

406 Danian, where values show a show a very minor increase from -9.3 to -9.1 (Fig. 2)

407

408 **Table 1.** Hf-Nd isotopic compositions. Depth interval represents the range from where the sample was taken. Reported depth is the
409 depth used to calculate the sample age and plot the data in Fig. 2. Depth and depth interval are reported in meters below the sea
410 floor (mbsf). *Ages were calculated using an age model built from the biostratigraphy report by Valicenti et al. (1993). Error accounts
411 for the standard error. $\Delta\varepsilon_{Hf}$ was calculated using the clay array correlation: $\Delta\varepsilon_{Hf} = \varepsilon_{Hf} - (0.78 * \varepsilon_{Nd} + 5.23)$ published in Bayon et al. (2016).
412 2□ is the standard deviation calculated for both ε_{Nd} and ε_{Hf}.

Sample	Depth interval (mbsf)	Depth (mbsf)	Age (Ma)*	$^{143}Nd/^{144}Nd$	ε_{Nd}	2σ	$^{176}Hf/^{177}Hf$	ε_{Hf}	2σ	$\varepsilon_{Nd(80)}$	$\varepsilon_{Hf(80)}$	$\Delta\varepsilon_{Hf(80)}$
O-A1/1	2145 - 50	2147	55	0.512149	-9.39	0.12	0.282647	-4.87	0.16	-8.45	-3.60	-2.24
O-A1/5	2235 - 45	2240	57	0.512132	-9.72	0.12	0.28267	-4.08	0.21	-8.76	-2.87	-1.27
O-A1/10	2346 - 55	2349	60	0.512138	-9.59	0.13	0.282667	-4.16	0.18	-8.66	-2.97	-1.45
O-A1/14	2439 - 45	2442	63	0.512158	-9.21	0.14	0.282707	-2.77	0.14	-8.36	-1.59	-0.30
O-A1/16	2481 - 90	2485	64	0.512164	-9.1	0.13	0.282716	-2.44	0.18	-8.17	-1.29	-0.15
O-A1/18	2529 - 35	2532	65	0.512159	-9.19	0.14	0.282707	-2.75	0.15	-8.30	-1.63	-0.39
O-A1/21	2562 - 71	2560	67	0.51215	-9.37	0.06	0.282715	-2.48	0.2	-8.47	-1.36	0.01
O-A1/23	2577 - 83	2580	72	0.512152	-9.33	0.16	0.282711	-2.63	0.18	-8.44	-1.49	-0.13
O-A1/24	2583 - 92	2588	73	0.512142	-9.51	0.14	0.282679	-3.74	1.8	-8.64	-2.62	-1.11
O-A1/27	2604 - 07	2605	74	0.512147	-9.42	0.17	0.282701	-2.96	0.2	-8.52	-1.86	-0.44
O-A1/30	2619 - 25	2622	76	0.51215	-9.37	0.15	0.282704	-2.88	0.24	-8.46	-1.73	-0.36
O-A1/33	2637 - 43	2640	78	0.512145	-9.47	0.11	0.282693	-3.26	0.26	-8.55	-2.16	-0.72
O-A1/37	2661 - 67	2665	80	0.512157	-9.22	0.17	0.282692	-3.3	0.25	-8.23	-2.09	-0.90
O-A1/37 (2)	2661 - 67	2665	80	0.512167	-9.04	0.12	0.282695	-3.18	0.17	-8.23	-2.09	-0.90
O-A1/40	2685 - 94	2690	83	0.512167	-9.04	0.13	0.282725	-2.14	0.29	-8.13	-1.01	0.11
O-A1/43	2790 - 96	2792	85	0.512172	-8.94	0.17	0.282695	-3.17	0.32	-7.98	-2.12	-1.12
O-A1/46	2943 - 46	2944	86	0.5122	-8.4	0.14	0.282696	-3.15	0.28	-7.42	-2.13	-1.57
O-A1/49	3102 - 08	3105	88	0.512214	-8.11	0.17	0.282704	-2.87	0.4	-7.14	-1.77	-1.44
O-A1/51	3201 - 07	3204	89	0.512201	-8.37	0.14	0.282667	-4.17	0.36	-7.45	-3.18	-2.59
O-A1/52	3255 - 61	3258	90	0.512208	-8.22	0.13	0.282668	-4.12	0.35	-7.32	-3.20	-2.73
O-A1/57	3492 - 98	3494	93	0.512214	-8.12	0.13	0.282685	-3.53	0.31	-7.14	-2.53	-2.19
O-A1/60	3582 - 91	3587	95	0.512206	-8.27	0.1	0.282661	-4.38	0.36	-7.31	-3.41	-2.94
O-A1/63	3624 - 27	3624	97	0.512183	-8.72	0.14	0.282693	-3.24	0.3	-7.73	-2.32	-1.52
O-A1/66	3693 - 96	3694	99	0.512132	-9.71	0.14	0.282698	-3.09	0.29	-8.63	-2.06	-0.56
O-A1/70	3792 - 98	3795	102	0.512175	-8.87	0.15	0.282694	-3.21	0.26	-7.84	-2.41	-1.53

4.3 Major and REE elements

Major oxides concentrations are reported in the supplementary table S5. The SiO_2 wt% is predominant among the samples with percentages between 24% and 82%. Second in abundance are CaO wt%, Fe_2O_3 wt%, and Al_2O_3 wt%, with concentrations reaching up to 28%, 19%, and 15%, respectively. Other major oxides like MgO wt%, K_2O wt%, and Na_2O wt%, are present in percentages lower than 4 wt%, whereas major oxides like TiO_2 wt%, MnO wt%, P_2O_5 wt%, Cr_2O_3 wt%, and NiO wt%, are percentages lower than 1 wt%.

REE elements for the clay fraction (< 2μm) are reported together with other trace elements in supplementary table S2. The REE concentrations for samples below sample O-A1/43 (below 2793 mbsf – 85 Ma) display a flat pattern with a very subtle depletion in HREE over LREE (Fig. 3). In contrast, samples in the upper part of the section (above 2793 mbsf – 85 Ma) show REE patterns with a slight depletion in LREE and MREE. Additionally, these samples show positive Eu anomalies (Eu/Eu* > 1; Supplementary table S2). Such anomalies are likely introduced due to undercorrected isobaric interferences associated to high Ba contents (>2000 ppm; Supplementary table S2)(e.g., Lidman et al., 2019; Zhu and Itoh, 2021), therefore Eu values were removed from the REE plot.

4.4 Transmission electron microscopy

Six samples evenly distributed along the section were analysed through TEM imagery (Fig. 4; Supplementary table S1). At the base of the section (sample O-A1/88; 4151mbsf – 112 Ma) clay particles show rather regular textures with defined edges, specially illite particles that are

present as elongated euhedral prisms (Fig. 4A). By the low-middle part of the section (sample O-A1/61; 3607 mbsf – 96 Ma) particles texture show a rather altered structure, and illite-smectite mixed-layers (I-S R_1) start to appear. The texture shows smaller, thinner, elongated and more fibrous particles (Fig. 4B; 'hairy-like' type). Above (sample O-A1/50; 3155 mbsf – 88 Ma), illite-smectite mixed-layers (I-S R_1) seem less altered, but still rather long and fibrous (Fig. 4C). Kaolinite also starts to become more frequent, with a characteristic hexagonal regular shaped texture (Fig. 4C). By the upper-middle part of the section (O-A1/32; above 2635 mbsf – 77 Ma), clay particles show predominantly flaky-shape texture, characteristic of smectite particles (Fig. 4D). Thin-elongated particles interpreted as illite also become more frequent (Fig. 4D).

5. Discussion

5.1 Clay minerals origin

Detrital clay minerals deposited in sedimentary basins are useful tracers to understand the evolution of surface continental processes (e.g., Bougeault et al., 2017; Chamley, 1989; Corentin et al., 2023, 2022; Dera et al., 2009; Gaitan et al., 2023). However, clay minerals can also form within sedimentary basins through processes such as hydrothermal activity, reverse weathering, alteration of volcanic ash falls, or in deep-sea conditions by the interaction between

sediments constituents and seawater (Chamley, 1989; Velde, 1995; Warr, 2022). Clay mineral assemblages can also be modified after formation or deposition during burial diagenesis in the basin (Chamley, 1989; Ketzer et al., 2003; Lanson et al., 2002). Therefore, to interpret variations in clay mineral assemblages in terms of surface continental processes, clay minerals at sedimentary basins should be identified first as detrital particles inherited from the landmasses where they were formed and without significant alteration.

The clay record of borehole O-A1 shows from top to bottom a clear and progressive influence of burial diagenesis, which is common for the depths range observed in borehole O-A1 (i.e., > 2500 mbsf; Fig. 2). At the top of the section, smectite predominates with minor presence of illite and chlorite-smectite mixed layers. Smectite disappears progressively with depth, illite-smectite (R_1) layers start to appear, and chlorite becomes more abundant in chlorite-smectite mixed layers, until only illite and chlorite are remaining at the base of the section (Fig. 2). This transition in clay assemblages is diagnostic of burial diagenesis. Smectites are the most sensitive clay minerals and start to transform into illite-smectite mixed layers around 70°C through illitization process, until a full transformation into illite is completed (Chamley, 1989; Lanson et al., 2009; Nadeau et al., 1985; Surdam and Crossey, 1987; Warr, 2022). Illitization and chloritization processes lead to the complete transformation into illite and/or chlorite depending on the chemistry of fluids and enclosed rocks, in particular the availability of K and Mg (Chamley, 1989; Lanson et al., 2009; Warr, 2022). The presence of kaolinite from the middle part of the section (ca. 3000-3600 mbsf) to the top can still represent a remnant of a detrital component, as kaolinite is often more resistant and preserved in diagenetically affected sediment deposits under 120-140 °C (Chamley, 1989; Lanson et al., 2002; Warr, 2022). The smectite peak at the very bottom of the section (ca. 4180 mbsf) is highly unusual at such depths. In this case, this peak is rather related to interlayered chlorite-smectite (i.e., corrensite). The presence of corrensite has been reported in metabasalts and

related to hydrothermal environments (Bettison-Varga, 1997; Inoue and Utada, 1991), which would be consistent in this case as the sediments at the bottom of the section are in contact with the basaltic basement that due to the depth and temperature could have undergone low-degree metamorphism.

The TEM images across the section support this interpretation (Fig. 4). Clay mineral textures in the upper part of the section (Campanian-Ypresian; 2650-2100 mbsf) indicate a rather detrital origin (Fig. 4D). The flake-shaped textures on smectite particles, and the polygonal shaped texture of illite and chlorite are indicative of detrital origin (Chamley, 1989; Clauer, 1990; Fagel et al., 2001). Below this upper part (2650 mbsf; Campanian), the progressive appearance of Illite-Smectite (R_1) particles with fibrous textures is already indicative of illitization, consistent with burial diagenetic processes (Bauluz et al., 2012; Lanson et al., 2009; Pearson and Small, 1988). Such textures become more prominent with depth, as interlayered particles become smaller, thinner, and occur as elongated fibres (i.e., 'hairy-like' textures — Fig. 4B-C), which is commonly observed in diagenetic affected sequences (Lanson et al., 2009; Nadeau et al., 1985). A semi-quantitative approach to interpret these clay mineral textures is the calculation of the Kübler or 'crystallinity' index on illite-smectite interstratified clay particles at the 10-Å X-ray diffraction peak. High crystallinity (i.e., low FWHM — Full Width at Half-Maximum height) is associated to clays recrystallised during diagenesis, whereas lower crystallinity (i.e., high FWHM) is associated to detrital non-altered clay particles (Kübler and Jaboyedoff, 2000; Jaboyedoff et al., 2001). This index shows the progressive effect of diagenesis between 2500 and 3800 mbsf, with a progressive increase in crystallinity along this interval of the section (FS2). The section above 2500 mbsf shows a shift to lower FWHM values, these values are associated to the absence of illite-smectite interstratified clay particles, which is also observed in the clay mineralogy with a significant increase in the smectite content above 2500 mbsf (Fig. 2). This level appears to represent the

514 threshold between the pristine detrital clay particles and the clay minerals altered by burial
515 diagenesis processes. Similarly, the values around 0.4 observed below 3800 msbf (FS2) seem
516 to be linked to a change in lithology in the borehole, shifting from claystones to more silty-
517 sandy sediments. Therefore, illite-smectite interstratified clay particles are rather absent in this
518 interval and their crystallinity cannot be assessed. REE concentrations can also offer insights
519 into the origin of clay minerals (Abbott et al., 2019; Bayon et al., 2023; Cullers et al., 1975).
520 Excluding the Eu/Eu* anomalies as they are likely related to undercorrected high Ba contents
521 in the samples (Fig. 3; see Major and REE elements 4.3) (e.g., Lidman et al., 2019; Zhu and
522 Itoh, 2021), the REE exhibit predominantly flat patterns, which are commonly observed in
523 detrital clays (Fagel et al., 1992; Zhao et al., 2021). Although there is some variability within
524 the REE patterns, with part of the samples showing a slight depletion in LREE, these patterns
525 resemble those observed in clay particles derived from large river basins (Bayon et al., 2015).
526 More importantly, the REE patterns do not display any Ce anomalies accompanied by marked
527 enrichments in HREE characteristic of seawater patterns, thus suggesting that authigenic
528 imprints during the early diagenesis stages are mostly absent (Nozaki, 2001; Zhao et al., 2021).
529 The burial diagenetic imprint observed on clay minerals does not appear either to have
530 impacted the REE concentrations. The clay mineral assemblages affected by burial diagenesis
531 from below the middle part of the section (2700 mbsf; Santonian) display in fact the most
532 typical shale-like flat patterns (Fig. 3).

533 The clay mineralogy, the clay particle textures, and the Kübler index indicate clearly
534 that the clay minerals assemblages, at least between 2500 and 3800 mbsf, have been modified
535 by burial diagenesis, although REE patterns seem to be unaffected. This interval within the
536 borehole corresponds to most of the late Cretaceous section, and therefore the changes in clay
537 mineralogy from borehole O-A1 cannot be interpreted in terms of changes in continental
538 denudation processes along the South African Plateau.

5.2 Sediment provenance

Sediment provenance in ancient environments has been largely assessed through neodymium isotopes (e.g., Goldstein et al., 1984; Hindshaw et al., 2018; Peucker-Ehrenbrink et al., 2010). This isotopic system is particularly suited for provenance studies because it is relatively unaffected by weathering processes (Goldstein and Jacobsen, 1988; Hindshaw et al., 2018; McCulloch and Wasserburg, 1978) and as Nd is a quite immobile element, it is resistant to diagenetic effects (e.g., Chakrabarti et al., 2007; Clauer and Chaudhuri, 1995). This is supported by the absence of correlation between $\varepsilon_{Nd(t)}$ and the Kübler Index (FS3), suggesting that burial diagenesis has not affected the evolution of ε_{Nd} values at this site.

The $\varepsilon_{Nd(80)}$ values from site O-A1 are relatively stable around -8.5 ε-units throughout the studied interval, except for a radiogenic excursion encompassing the Cenomanian to Coniacian interval where values reach about -7. The recorded clay $\varepsilon_{Nd(80)}$ values are compatible with a mixed contribution of different sediment sources located on the western-central areas of the South African Plateau (Fig. 5; Gaitan et al., 2023). The increase from -8.6 to -7.1 observed in clay $\varepsilon_{Nd(80)}$ values between the Cenomanian to Coniacian interval (ca. 97-87 Ma) points to an increase in sediment input from rather radiogenic sources (Fig. 6). Such contribution could have come from the Saldania Belt located in the southwestern African margin (Chemale et al., 2011; mean $\varepsilon_{Nd(80)}$ = -6; Supplementary table S6) or the Karoo basalts (Hawkesworth et al.,

1984; mean $\varepsilon_{Nd(80)}$ = -4.8; Supplementary table S6). A concomitant increase in P_2O_5 wt% and clay $\varepsilon_{Nd(80)}$ values during the Cenomanian to Coniacian interval tends to support contributions from the Karoo basalts rather than the Saldania Belt (Fig. 6). P_2O_5 wt% content is typically high (> 0.4 wt%) in volcanic rocks, including Continental Flood Basalts (CFB) like the Karoo basalts (e.g., Carlson, 1991; Jourdan et al., 2007). From the Coniacian onwards, the clay $\varepsilon_{Nd(80)}$ values decrease, reaching values around -8.5 in the Campanian (ca. 80 Ma), and remaining stable up to the Ypresian (Fig. 6). These more negative values indicate increased contributions from more unradiogenic units (i.e., lower ε_{Nd} values), such as the Namaqua Belt (Reid, 1997; mean $\varepsilon_{Nd(80)}$ = -15.7; Supplementary table S6), the Cape Belt (Dia et al., 1990; mean $\varepsilon_{Nd(80)}$ = -10; Supplementary table S6) or the Karoo sediments (Dia et al., 1990; mean $\varepsilon_{Nd(80)}$ = -8.9; Supplementary table S6), all located at southwestern – central areas of the South African Plateau (Fig. 5). Given the nature of these units which include very heterogeneous lithologies, further analyses including trace elements and other isotopic systems will be required to better constrain their relative contribution to the sediment sources at site O-A1.

The changes in contribution from the sediment sources presented here can be partially associated to the tectonic uplift experienced by the South African Plateau throughout the late Cretaceous. The tectonic uplift of the Plateau involved a margin tilting, with an early uplift

phase (ca. 93-80 Ma) affecting predominantly the eastern margin, followed by a second phase (ca. 80-70 Ma) affecting predominantly the western margin (Baby et al., 2019, 2018a; Braun et al., 2014; Stanley et al., 2021). This tilting has been inferred based on changes in sedimentation rates from the basins adjacent to the south African margin (Baby et al., 2019, 2018a; Braun et al., 2014; Stanley et al., 2021). Although the onset of the early uplift stage is set around 93 Ma (Baby et al., 2019, 2018a; Braun et al., 2014; Stanley et al., 2021), thermochronological data reports erosional cooling as early as 100 Ma (e.g., Flowers and Schoene, 2010; Stanley et al., 2013; Wildman et al., 2015). This eastern margin uplift could have brought sediments from central areas of the Plateau to the Cape Basin, which could be consistent with the increased input of radiogenic sediments derived from the Karoo basalts. Similarly, the switch to more unradiogenic clay $\varepsilon_{Nd(80)}$ values after the Coniacian (ca. 87 Ma) could be related to the input of sediments from unradiogenic units (e.g., Namaqua, Karoo sediments or Cape Belt), which could have been progressively incorporated during the later phase of this tectonic uplift stage (ca. 93-80 Ma; Fig. 6). The $\varepsilon_{Nd(80)}$ values stabilise and remain quite stable around -8.5 after the early Campanian (ca. 80 Ma), suggesting that the sediment sources contributions did not change during the later uplift stage of the South African Plateau (ca. 80-70 Ma).

Along the South African Plateau a main drainage reconfiguration has been identified around the Maastrichtian (ca. 70 Ma), suggesting that the Kalahari and Karoo paleorivers merged into the modern Orange river system due to tectonic activity (De Wit et al., 2009; De Wit, 1999; Goudie, 2005). Our data suggests that previous reconfigurations might have occurred and could explain the shift between the contribution of the Karoo basalts — recorded

by the radiogenic $\varepsilon_{Nd(80)}$ excursion and the associated higher P_2O_5 wt% content — to a more relevant contribution of sediments derived from the Namaqua belt, Karoo sediments, and Cape Belt, recorded by the decrease in $\varepsilon_{Nd(80)}$ during the Coniacian-Campanian interval. Unlike the early uplift stage, the late uplift stage (ca. 80-70 Ma) of the western margin does not seem to have had a major impact in the sediment sources contribution, as by the Campanian (ca. 80 Ma), the $\varepsilon_{Nd(80)}$ values have already stabilised and remained quite stable up to the Ypresian (Fig. 5 and 6). This stability in the values also indicate that the suggested drainage reconfiguration around the Maastrichtian (ca. 70 Ma) (De Wit et al., 2009; De Wit, 1999; Goudie, 2005) did not have an impact in sediment sources, only in sediments distribution in the Cape Basin. This is observed in the contrasting changes in sediment fluxes observed between the Orange and Cape Basins (Fig. 6), both located adjacent to the southwestern African margin.

5.3 Evolution of silicate chemical weathering in the southwestern African margin

The evolution of silicate chemical weathering is often investigated using major element ratios and indices (e.g., Baiyegunhi et al., 2017; Dinis et al., 2020). Among these indices the most commonly used is the CIA index (Chemical Index of Alteration; Nesbitt and Young, 1982). However, at site O-A, the impact of burial diagenesis across the section could have perturbed the index, especially as Na and K are very mobile elements and could have been impacted by illitization processes. In addition, this index is sensitive to sediment sorting and

changes in sediment sources, which vary at this site as indicated by changes in lithology and in clay $\varepsilon_{Nd(80)}$ values. Hence, the use of the CIA is not adequate for site O-A1. To investigate the evolution of silicate chemical weathering, we employed the combined Sm-Nd and Lu-Hf isotopic systems from clay fractions, which have already been used in both modern and ancient environments and have been shown to be independent to changes in sediment sources (e.g., Bayon et al., 2012, 2016, Corentin et al., 2023). The clay ε_{Nd} and ε_{Hf} values are not correlated to the Kübler index (FS3), suggesting that the burial diagenetic processes identified in this site did not affect the clay isotopic compositions. During magmatic processes, the Sm-Nd and Lu-Hf isotope systems behave alike, with mantellic material enriched in Sm and Lu, leading with time to more radiogenic ε_{Nd} and ε_{Hf} values in mantellic material when compared to crustal material. This is highlighted by the correlation of crustal and mantellic ε_{Nd} and ε_{Hf} material, called the "Terrestrial Array" (Vervoort et al., 2011). Conversely, during surface continental processes these isotope systems behave differently, leading to a decoupling between ε_{Nd} and ε_{Hf} in weathering products. Autogenic processes within sediment transport such as mineral sorting are partly responsible for this decoupling, mainly due to the control exerted by zircon over ε_{Hf} values, as zircon is characterized by high concentrations and highly unradiogenic Hf values (Bayon et al., 2006; Carpentier et al., 2014; Patchett et al., 1984). In consequence, the

clay fraction — usually devoid of zircons in comparison to coarser fractions — displays higher ε_{Hf} values (Carpentier et al., 2014; Chauvel et al., 2014; Garçon et al., 2013). The clay fraction of sediments display a distinct correlation between ε_{Nd} and ε_{Hf} values denominated the "Clay Array" (Bayon et al., 2016). Within the clay fraction, it has been shown that the offset in ε_{Hf} values from the clay array, expressed as $\Delta\varepsilon_{Hf}$, is dependent on silicate weathering intensity (Bayon et al. 2016). Enhanced weathering would tend to augment the release of radiogenic Hf from easy weatherable and Lu-enriched mineral phases (Bayon et al., 2006) into the solutes from which clay minerals precipitate, therefore they exhibit an increase in their ε_{Hf} values.

While zircons are typically present in trace amounts within clay fractions, if they are present in substantial quantities they can have an impact on the evolution of ε_{Hf} on clay fractions. In our study, Zr concentrations of the analyzed samples oscillate between 120 and 180 ppm (Supplementary table S2), which falls within the range of the zircon-poor clay fraction and thus discards any significative influence of zircons in our ε_{Hf} values (Bayon et al., 2016; Corentin et al., 2022). Most importantly, there is no correlation between Zr concentrations and the $\Delta\varepsilon_{Hf}$ and ε_{Hf} values (FS4), indicating that changes in $\Delta\varepsilon_{Hf}$ are not controlled by zircon abundance, but can be attributed mainly to changes in silicate chemical weathering intensity.

Following Equation 1, $\Delta\varepsilon_{Hf(80)}$ values were calculated to assess changes in silicate chemical weathering intensity. As for ε_{Nd} values, an age correction of t = 80 Ma has been uniformly applied to the ε_{Hf} values to calculate the $\Delta\varepsilon_{Hf(80)}$ (Table 1). This correction corresponds to the average age of the analysed time interval. Based on Bayon et al. (2016), the increase in $\Delta\varepsilon_{Hf(80)}$ values observed from the Cenomanian to the Maastrichtian (ca. 95-68 Ma; Fig. 6) points to a long-term enhancement of silicate chemical weathering intensity in the South African Plateau, that only decreases after the Cretaceous-Paleogene (K-Pg) boundary. The

initial increase in $\Delta\varepsilon_{Hf(80)}$ values between the Cenomanian and the Coniacian is concomitant to the suggested weathering of the Karoo basalts, as previously shown by $\varepsilon_{Nd(80)}$ values and P_2O_5 wt%. However, while there is a shift to more unradiogenic $\varepsilon_{Nd(80)}$ values around the Coniacian (ca. 87 Ma), indicating the input of a different sediment source, the $\Delta\varepsilon_{Hf(80)}$ still shows a slight increase in the intensity of silicate chemical weathering that extends up to the Maastrichtian.

The new combined ε_{Nd} and ε_{Hf} data from site O-A1 support the enhancement in silicate chemical weathering processes in the South African Plateau during the late Cretaceous, which has been depicted at the more distal site DSDP 361 (Gaitan et al., 2024). Both sites display mostly concordant sediment sources as suggested by the $\varepsilon_{Nd(80)}$ values (Fig. 6). However, the geochemical signals recorded at both sites display differences in their evolution. While records from borehole O-A1 show an enhancement in silicate chemical weathering through an increase in $\Delta\varepsilon_{Hf(80)}$ values between the Cenomanian to the Maastrichtian (Fig. 6), records from DSDP core 361 show an increase from the Campanian to the Danian. Part of this discrepancy can be related to the uncertainties in the age model from DSDP core 361, which are more pronounced for the Turonian-Santonian interval (Gaitan et al., 2023). However, during the Maastrichtian-Danian interval where the age model is better calibrated for both sites their $\Delta\varepsilon_{Hf(80)}$ records clearly differ. At borehole O-A1, the decrease in chemical weathering starts at the K-Pg boundary, whereas at DSDP core 361 $\Delta\varepsilon_{Hf(80)}$ continues to increase and only begins to decrease at the end of the Danian (ca. 58 Ma; Fig. 6). This time lag is also observed in the $\varepsilon_{Nd(80)}$ values. Clay $\varepsilon_{Nd(80)}$ values from borehole O-A1 remain rather stable from the Campanian upwards whereas clay $\varepsilon_{Nd(80)}$ values at DSDP core 361 display a pronounced decrease of about 2 ε-units in the Maastrichtian-Danian interval, increasing again during the Danian-Thanetian interval (Fig. 6). These differences in the isotopic record during the K-Pg transition could be related to the input of different source material into the abyssal plain where the DSDP core 361 is located, which would explain differences in $\varepsilon_{Nd(80)}$ values between the two sites at that time. As the

DSDP core 361 is located in a distal area, it has already been suggested that deep-bottom currents could have brought allogenic sediments from afar to this site (Murphy and Thomas, 2013; Natland, 1978). Likewise, aeolian transport of fine particles could incorporate allogenic material to distal areas. Overall, all these factors could have contributed to a divergence between borehole O-A1 and DSDP core 361 sources.

5.4 Insights on climate, erosion, and weathering.

Clay mineralogy is commonly used to assess the relative evolution of physical erosion (e.g., Corentin et al., 2023, 2022; John et al., 2012), however, the clay mineralogy from site O-A1 cannot be used. The clay assemblages have been clearly affected by burial diagenetic processes at least below 2500 mbsf (ca. 66 Ma), thus their content is not indicative of surface continental processes below this depth. Published clay mineral assemblages reported for the abyssal plain of the Cape Basin (i.e., DSDP 361) show an increase in illite and chlorite over smectite during the Campanian-Danian interval (Gaitan et al., 2023). This increase in primary clays over secondary clays has been interpreted as a relative increase in physical erosion processes, and it is coherent with existing thermochronological analyses across the South African Plateau that have constrained rock cooling episodes interpreted as enhanced denudation during the mid-late Cretaceous (Brown et al., 2002; Gallagher and Brown, 1999; Green et al., 2017; Kounov et al., 2009, 2013; Stanley et al., 2013, 2015; Stanley and Flowers, 2020; Tinker et al., 2008; Wildman et al., 2015, 2016). The onset of these cooling episodes around the margins — interpreted primarily as erosional cooling — are dated around 100 Ma and extending up to 70 Ma (Brown et al., 2002; Gallagher and Brown, 1999; Green et al., 2017; Kounov et al., 2009, 2013; Tinker et al., 2008; Wildman et al., 2015, 2016). Additional AHe data (Apatite-Helium)

from inner areas of the Plateau have also shown an eastern erosional wave between ca. 120 and 60 Ma (Stanley et al., 2015, 2013; Stanley and Flowers, 2020).

These rock cooling episodes have been used to link tectonic uplift with enhanced physical erosion and increases in sedimentation rates along the south African Margin during the late Cretaceous (Baby et al., 2019, 2018a; Braun et al., 2014; Stanley et al., 2021). This period also corresponds to the main increase in $\Delta\varepsilon_{Hf(80)}$ associated to enhanced weathering of the margin, suggesting that the tectonic uplift of the South African Plateau during the late Cretaceous has resulted in an enhancement of both physical erosion and chemical weathering. The coupled response of both processes suggests that denudation in this area likely operated under transport-limited conditions (Gabet and Mudd, 2009; West et al., 2005), in which erosion and weathering are positively correlated. This transport-limited regime has been recently suggested to be optimal for CO_2 withdrawal through silicate reactions (Bufe et al., 2024), particularly because in these settings chemical weathering reactions are not kinetically-limited and therefore the intensity of silicate chemical weathering reactions is not limited or reduced (Caves Rugenstein et al., 2019; Gabet and Mudd, 2009; Maher and Chamberlain, 2014).

The increase in silicate chemical weathering recorded by $\Delta\varepsilon_{Hf(80)}$ data starting by the Cenomanian-Turonian is concomitant to the onset of the global climatic cooling recorded during the late Cretaceous (O'Brien et al. 2017). This increase during cooling climate conditions hints to tectonics over climate as the main driver of weathering processes, as cooler conditions tend to hamper the kinetics of silicate chemical weathering reactions (Maher, 2010; Riebe et al., 2004). More importantly, this enhanced weathering episode could in contrast have contributed to the onset of this cooling trend via silicate reactions. In fact, other sites on the West and South African margin (Corentin et al., 2024; Gaitan 2023), and on the eastern South American margin (Corentin et al., 2023, 2022), have also recorded increases in silicate

734 chemical weathering, which in spite starting around the Santonian, have been suggested to have
735 contributed to the long-term global climate trend recorded during the late Cretaceous.
736
737 The increase in $\Delta\varepsilon_{Hf(80)}$ in site O-A1 also precedes an increase in Ba (ppm) and CaO wt%
738 during the Santonian-Maastrichtian interval (ca. 85-65 Ma) suggesting also an increase in
739 marine productivity (Fig. 7A). The increase in CaO wt% could be initially linked to carbonate
740 productivity, especially due to the presence of the planktonic foraminifera used to establish the
741 age model of the site. However, to determine whether this increase is associated to marine
742 productivity or to a detrital input, the fraction of Ba can be normalised to Al to determine the
743 biogenic component of Ba (Ba_{xs}), which can be used as a proxy to determine export production
744 (i.e., carbon flux to the deep ocean) (Bridgestock et al., 2019; Dymond et al., 1992; Eagle et
745 al., 2003; Liu et al., 2018; Paytan and Griffith, 2007). The evolution of Ba_{xs} shows indeed an
746 increase during the Santonian-Maastrichtian interval, but more importantly appears to show a
747 positive correlation with $\Delta\varepsilon_{Hf(80)}$ (Fig. 7B), suggesting that enhanced silicate chemical
748 weathering could represent a source of nutrients that could have promoted and sustained high
749 productivity in this setting (e.g., Pogge von Strandmann et al., 2013; Yobo et al., 2021). This
750 correlation between increases in silicate chemical weathering based on $\Delta\varepsilon_{Hfclays}$ and export
751 production, has already been proposed as a mechanism in which silicate chemical weathering
752 can impact atmospheric CO_2 levels through enhancement of organic carbon burial by increases
753 in nutrient release, in addition to the CO_2 drawdown from silicate weathering reactions (Chen
754 et al., 2023). This hypothesis however, needs to be further confirmed by additional analyses
755 such as TOC (total organic content), $\delta^{13}C$, or organic phosphorus, that could give independent
756 insights on the potential evolution of export production (Schoepfer et al., 2015).
757 Our new data from site O-A1 reveals an onset of enhanced silicate chemical weathering as
758 early as the Cenomanian (ca. 95 Ma; Fig. 6), also potentially contributing to promote and

sustain primary productivity between the Santonian and the Maastrichtian (Fig. 7). This suggests, for the first time, that the increase in silicate chemical weathering across the South African Plateau could have contributed as one of the triggering factors in the atmospheric CO_2 withdrawal and subsequent onset of the global cooling trend during the late Cretaceous. First, through atmospheric CO_2 withdrawal by silicate weathering reactions, and later in the Santonian - Maastrichtian by potentially enhancing primary productivity and organic carbon storage sustained by enhanced nutrient supply from the South African continent. Still, coupled geochemical-climate models are needed to constrain quantitatively the impact of these processes in atmospheric CO_2 withdrawal and on the global climate.

6. Conclusion

Our data from borehole O-A1 suggest, for the first time, that tectonically driven enhancement of silicate chemical weathering along the South African Plateau might have played a crucial role in the triggering of the late Cretaceous global cooling trend that started around the Cenomanian-Turonian transition. Additionally, the impact of enhanced silicate chemical weathering in CO_2 seems to extend beyond the silicate mineral reactions, and also could be potentially related to the enhancement of primary productivity and organic carbon storage, which is a process that also captures carbon.

Our new $\Delta\varepsilon_{Hf(80)}$ data indicate an increase in silicate chemical weathering from the Cenomanian up to the Maastrichtian. The early phase (i.e., Cenomanian-Coniacian) of enhanced erosion and silicate chemical weathering is linked to the denudation of the Karoo basalts. This is supported by $\varepsilon_{Nd(80)}$ and P_2O_5 wt% data, which show patterns relatable to the incorporation of sediments derived from basaltic volcanic rocks. The $\varepsilon_{Nd(80)}$ data suggest that the main sediment source of the Cape Basin are units from the western-central areas of the South African Plateau, including the Karoo basalts, Karoo sediments, and potentially the Cape, Saldania, and Namaqua belts. Yet, there are some discrepancies between the DSDP core 361 and the borehole O-A1 record. Discrepancies in the $\Delta\varepsilon_{Hf(80)}$ evolution during the Cenomanian-Santonian interval (ca. 97-83 Ma) are likely linked to age model uncertainties from the DSDP core 361. However, discrepancies during the Maastrichtian-Thanetian interval appear to indicate that drainage reconfiguration and changes in sediment distribution and source, altered the denudation record between both locations during this period. Overall, our study highlights the pertinence of extended basin analysis using multiple records across the basin to understand

and interpret denudation records, and to link them with changes in response to climatic and tectonic forcings.

Figure Captions

Figure 1. Map of the source-to-sink system composed by the South African plateau and the Cape basin. O-A1 borehole (33.161539°S; 16.822406°E) is located at the continental slope of the Cape Basin. DSDP core 361 is located in the abyssal plain of the basin (35.066167°S; 15.448500°E). Topography and bathymetry data compiled from NASA JPL, (2013).

Figure 2. Biostratigraphy (planktonic foraminifera), sedimentary log, bulk mineralogy, clay assemblages, and present-day □$_{Nd}$-□$_{Hf}$ isotope compositions of the analysed samples from borehole O-A1. Units refer to the classification established by Baby et al., (2019). Planktonic foraminifera abbreviations: *R. appenninica=Rotalipora appenninica*; *H. helvetica=Helvetoglobotruncana helvetica*; *M. coronata=Marginotruncana coronata*; *G. elevata = Globotruncanita elevata; G. aegyptiaca = Globotruncanita aegyptiaca; A. mayaroensis = Abathomphalus mayaroensis*. Error bars represent the standard deviation (2□) calculated for both ε_{Nd} and ε_{Hf}. Both ε_{Nd} and ε_{Hf} isotopic values are reported for t = 0.

Figure 3. REE concentrations of the clay fraction (< 2□m) from borehole O-A1. REE contents were normalized to the Post-Archean average Australian Shale (PAAS; Taylor and McLennan, 1985).

Figure 4. A) TEM image from sample O-A1/88 exhibiting long elongated regular-shaped illite particles. **B)** TEM image from sample O-A1/61 showing interlayered illite-smectite highly altered, thin, and fibrous. **C)** TEM image from sample O-A1/50 showing interlayered illite-

816 smectite, showing an elongated fibrous texture. At the bottom of the image a hexagonal
817 kaolinite particle can be observed as well. **D)** TEM image from sample O-A1/32 showing
818 smectite particles and regular polygonal illite and chlorite particles.

819 **Figure 5**. Lithological units from the South African Plateau. The ε_{Nd} data compilation are as

820 follow: 1) Karoo Basalts (Hawkesworth et al., 1984) 2) Cape Belt and Karoo Sediments (Dia
821 et al., 1990) 3) Namaqua Belt (Reid, 1997) 4) Saldania Belt (Chemale et al., 2011). Modified
822 after Gaitan et al. (2023).

823 **Figure 6.** P_2O_5 wt%, $\varepsilon_{Nd(80)}$ and $\varepsilon_{Hf(80)}$ values for borehole O-A1 (continental slope), $\varepsilon_{Nd(80)}$

824 and $\varepsilon_{Hf(80)}$ values are also reported for the DSDP 361 core (abyssal plain; Gaitan et al., 2023).
825 Values were calculated at t = 80 Ma. Palaeoprecipitations of the South African plateau are
826 based on flora data set (Rayner et al., 1997; Sandersen, 2006; Smith, 1986). Sedimentation
827 rates of the Cape and Orange Basins are estimations done by Baby et al. (2019). $\delta^{18}O$ records
828 from global benthic foraminifera are compiled from Friedrich et al. (2012); including data from
829 Zachos et al. (2008) for the Cenozoic. Sample O-A1/37 was the only sample with a replicate
830 for both the ε_{Nd} and ε_{Hf} values, the values were averaged, and the mean was used for the

831 plots. A smoothed curve and its 95% confidence interval were calculated for the P_2O_5 wt%
832 using Kernel regressions **1)** Rock cooling interval based on multiple AFT and AHe
833 thermochronology data across the South African Plateau (Brown et al., 2002; Gallagher and
834 Brown, 1999; Green et al., 2017; Kounov et al., 2009, 2013; Raab et al., 2002; Stanley et al.,
835 2013, 2015, 2015; Stanley and Flowers, 2020; Tinker et al., 2008; Wildman et al., 2015, 2016)

2) Timing of the South African Plateau margins uplift is based on the erosion-deposition model proposed by Baby et al. (2019).

Figure 7. A) Evolution of Ba (ppm) and CaO wt% along the borehole O-A1. **B)** Linear correlation and its 95% confidence interval between Ba_{xs} and $\varepsilon_{Hf(80)}$. Ba_{xs} was calculated as: $Ba_{xs} = Ba_{total} - \left(\frac{Ba}{Al}\right)_{ter} \times Al_{total}$. $(Ba/Al)_{ter}$ ratio value used for the calculations was 0.0075, which reflects the average upper crust (Dymond et al., 1992).

FS1. Sedimentary log of borehole O-A1 and the age model used to determine the ages along the section. Planktonic foraminifera were used to build the age model. Abbreviations: M. coronata = Marginotruncana coronata. Species used to define the anchor points (white circles) of the age model are shown on the graph.

FS2. Kûbler index of crystallinity along the borehole O-A1 with the evolution on lithology and clay assemblages.

FS3. Kûbler index compared to the evolution of $\varepsilon_{Nd,}$ ε_{Hf}, and $\varepsilon\varepsilon_{Hf}$. No correlation is found between the index and the isotope measurements.

FS4. ε_{Hf} vs Zr (in ppm) and $\varepsilon\varepsilon_{Hf}$ vs Zr (in ppm). Zr content is assumed as an indicator of the presence of zircon. The R^2 coefficient for both graphs shows low correlation between the values.

References

Abbott, A.N., Löhr, S., Trethewy, M., 2019. Are Clay Minerals the Primary Control on the Oceanic Rare Earth Element Budget? Front. Mar. Sci. 6.

858 Aizawa, M., Bluck, B., Cartwright, J., Milner, S., Swart, R., Ward, J., 2000. Constraints on the
859 geomorphological evolution of Namibia from the offshore stratigraphic record.
860 Baby, G., Guillocheau, F., Boulogne, C., Robin, C., Dall'Asta, M., 2018a. Uplift history of a
861 transform continental margin revealed by the stratigraphic record: The case of the
862 Agulhas transform margin along the Southern African Plateau. Tectonophysics 731–
863 732, 104–130. https://doi.org/10.1016/j.tecto.2018.03.014
864 Baby, G., Guillocheau, F., Braun, J., Robin, C., Dall'Asta, M., 2019. Solid sedimentation rates
865 history of the Southern African continental margins: Implications for the uplift history
866 of the South African Plateau. Terra Nova 32, 53–65. https://doi.org/10.1111/ter.12435
867 Baby, G., Guillocheau, F., Morin, J., Ressouche, J., Robin, C., Broucke, O., Dall'Asta, M.,
868 2018b. Post-rift stratigraphic evolution of the Atlantic margin of Namibia and South
869 Africa: Implications for the vertical movements of the margin and the uplift history of
870 the South African Plateau. Mar. Pet. Geol. 97, 169–191.
871 https://doi.org/10.1016/j.marpetgeo.2018.06.030
872 Baiyegunhi, C., Liu, K., Gwavava, O., 2017. Geochemistry of sandstones and shales from the
873 Ecca Group, Karoo Supergroup, in the Eastern Cape Province of South Africa:
874 Implications for provenance, weathering and tectonic setting. Open Geosci. 9, 340–360.
875 https://doi.org/10.1515/geo-2017-0028
876 Barrat, J.A., Keller, F., Amossé, J., Taylor, R.N., Nesbitt, R.W., Hirata, T., 1996.
877 DETERMINATION OF RARE EARTH ELEMENTS IN SIXTEEN SILICATE
878 REFERENCE SAMPLES BY ICP-MS AFTER TM ADDITION AND ION
879 EXCHANGE SEPARATION. Geostand. Geoanalytical Res. 20, 133–139.
880 https://doi.org/10.1111/j.1751-908X.1996.tb00177.x
881 Bauluz, B., Yuste, A., Mayayo, M.J., Rodríguez-Navarro, A.B., González-López, J.M., 2012.
882 Microtexture and genesis of clay minerals from a turbiditic sequence in a Southern

Pyrenees foreland basin (Jaca basin, Eocene). Clay Miner. 47, 303–318. https://doi.org/10.1180/claymin.2012.047.3.02

Bayon, G., Barrat, J., Etoubleau, J., Benoit, M., Bollinger, C., Révillon, S., 2009. Determination of Rare Earth Elements, Sc, Y, Zr, Ba, Hf and Th in Geological Samples by ICP-MS after Tm Addition and Alkaline Fusion 12.

Bayon, G., German, C.R., Boella, R.M., Milton, J.A., Taylor, R.N., Nesbitt, R.W., 2002. An improved method for extracting marine sediment fractions and its application to Sr and Nd isotopic analysis. Chem. Geol. 187, 179–199. https://doi.org/10.1016/S0009-2541(01)00416-8

Bayon, G., Giresse, P., Chen, H., Rouget, M.-L., Gueguen, B., Moizinho, G.R., Barrat, J.-A., Beaufort, D., 2023. The Behavior of Rare Earth Elements during Green Clay Authigenesis on the Congo Continental Shelf. Minerals 13, 1081. https://doi.org/10.3390/min13081081

Bayon, G., Skonieczny, C., Delvigne, C., Toucanne, S., Bermell, S., Ponzevera, E., André, L., 2016. Environmental Hf–Nd isotopic decoupling in World river clays. Earth Planet. Sci. Lett. 438, 25–36. https://doi.org/10.1016/j.epsl.2016.01.010

Bayon, G., Vigier, N., Burton, K.W., Jean Carignan, A.B., Etoubleau, J., Chu, N.-C., 2006. The control of weathering processes on riverine and seawater hafnium isotope ratios. Geology 34, 433. https://doi.org/10.1130/G22130.1

Berner, R.A., Lasaga, A.C., Garrels, R.M., 1983. Carbonate-silicate geochemical cycle and its effect on atmospheric carbon dioxide over the past 100 million years. Am J Sci U. S. 283:7. https://doi.org/10.2475/ajs.283.7.641

Bettison-Varga, L., 1997. The Role of Randomly Mixed-Layered Chlorite/Smectite in the Transformation of Smectite to Chlorite. Clays Clay Miner. 45, 506–516. https://doi.org/10.1346/CCMN.1997.0450403

908 Blichert-Toft, J., Chauvel, C., Albarède, F., 1997. Separation of Hf and Lu for high-precision
909 isotope analysis of rock samples by magnetic sector-multiple collector ICP-MS.
910 Contrib. Mineral. Petrol. 127, 248–260. https://doi.org/10.1007/s004100050278
911 Bougeault, C., Pellenard, P., Deconinck, J.-F., Hesselbo, S.P., Dommergues, J.-L., Bruneau,
912 L., Cocquerez, T., Laffont, R., Huret, E., Thibault, N., 2017. Climatic and
913 palaeoceanographic changes during the Pliensbachian (Early Jurassic) inferred from
914 clay mineralogy and stable isotope (C-O) geochemistry (NW Europe). Glob. Planet.
915 Change 149, 139–152. https://doi.org/10.1016/j.gloplacha.2017.01.005
916 Bouvier, A., Vervoort, J.D., Patchett, P.J., 2008. The Lu–Hf and Sm–Nd isotopic composition
917 of CHUR: Constraints from unequilibrated chondrites and implications for the bulk
918 composition of terrestrial planets. Earth Planet. Sci. Lett. 273, 48–57.
919 https://doi.org/10.1016/j.epsl.2008.06.010
920 Braun, J., Guillocheau, F., Robin, C., Baby, G., Jelsma, H., 2014. Rapid erosion of the Southern
921 African Plateau as it climbs over a mantle superswell: ERODING SOUTHERN
922 AFRICAN PLATEAU. J. Geophys. Res. Solid Earth 119, 6093–6112.
923 https://doi.org/10.1002/2014JB010998
924 Bridgestock, L., Hsieh, Y.-T., Porcelli, D., Henderson, G.M., 2019. Increased export
925 production during recovery from the Paleocene–Eocene thermal maximum constrained
926 by sedimentary Ba isotopes. Earth Planet. Sci. Lett. 510, 53–63.
927 https://doi.org/10.1016/j.epsl.2018.12.036
928 Brown, R.W., Summerfield, M.A., Gleadow, A.J.W., 2002. Denudational history along a
929 transect across the Drakensberg Escarpment of southern Africa derived from apatite
930 fission track thermochronology. J. Geophys. Res. Solid Earth 107, ETG 10-1-ETG 10-
931 18. https://doi.org/10.1029/2001JB000745

Bufe, A., Rugenstein, J.K.C., Hovius, N., 2024. CO2 drawdown from weathering is maximized at moderate erosion rates. Science 383, 1075–1080. https://doi.org/10.1126/science.adk0957

Carlson, R.W., 1991. Physical and chemical evidence on the cause and source characteristics of flood basalt volcanism. Aust. J. Earth Sci. 38, 525–544. https://doi.org/10.1080/08120099108727989

Carpentier, M., Weis, D., Chauvel, C., 2014. Fractionation of Sr and Hf isotopes by mineral sorting in Cascadia Basin terrigenous sediments. Chem. Geol. 382, 67–82. https://doi.org/10.1016/j.chemgeo.2014.05.028

Caves Rugenstein, J.K., Ibarra, D.E., von Blanckenburg, F., 2019. Neogene cooling driven by land surface reactivity rather than increased weathering fluxes. Nature 571, 99–102. https://doi.org/10.1038/s41586-019-1332-y

Chakrabarti, R., Abanda, P.A., Hannigan, R.E., Basu, A.R., 2007. Effects of diagenesis on the Nd-isotopic composition of black shales from the 420 Ma Utica Shale Magnafacies. Chem. Geol. 244, 221–231. https://doi.org/10.1016/j.chemgeo.2007.06.017

Chamley, H., 1989. Clay Sedimentology. Springer Science & Business Media.

Chauvel, C., Garçon, M., Bureau, S., Besnault, A., Jahn, B., Ding, Z., 2014. Constraints from loess on the Hf–Nd isotopic composition of the upper continental crust. Earth Planet. Sci. Lett. 388, 48–58. https://doi.org/10.1016/j.epsl.2013.11.045

Chemale, F., Scheepers, R., Gresse, P.G., Van Schmus, W.R., 2011. Geochronology and sources of late Neoproterozoic to Cambrian granites of the Saldania Belt. Int. J. Earth Sci. 100, 431–444. https://doi.org/10.1007/s00531-010-0579-1

Chen, H., Bayon, G., Xu, Z., Li, T., 2023. Hafnium isotope evidence for enhanced weatherability at high southern latitudes during Oceanic Anoxic Event 2. Earth Planet. Sci. Lett. 601, 117910. https://doi.org/10.1016/j.epsl.2022.117910

957 Clauer, N., 1990. Morphological, Chemical, and Isotopic Evidence for an Early Diagenetic
958 Evolution of Detrital Smectite in Marine Sediments. Clays Clay Miner. 38, 33–46.
959 https://doi.org/10.1346/CCMN.1990.0380105
960 Clauer, N., Chaudhuri, S., 1995. Clays in Crustal Environments. Springer Berlin Heidelberg,
961 Berlin, Heidelberg. https://doi.org/10.1007/978-3-642-79085-0
962 Cohen, K.M., Finney, S.C., Gibbard, P.L., Fan, J.-X., 2013. The ICS International
963 Chronostratigraphic Chart. Episodes J. Int. Geosci. 36, 199–204.
964 https://doi.org/10.18814/epiiugs/2013/v36i3/002
965 Collier, J.S., McDermott, C., Warner, G., Gyori, N., Schnabel, M., McDermott, K., Horn,
966 B.W., 2017. New constraints on the age and style of continental breakup in the South
967 Atlantic from magnetic anomaly data. Earth Planet. Sci. Lett. 477, 27–40.
968 https://doi.org/10.1016/j.epsl.2017.08.007
969 Corentin, P., Pucéat, E., Pellenard, P., Freslon, N., Guiraud, M., Blondet, J., Adatte, T., Bayon,
970 G., 2022. Hafnium-neodymium isotope evidence for enhanced weathering and uplift-
971 climate interactions during the Late Cretaceous. Chem. Geol. 591, 120724.
972 https://doi.org/10.1016/j.chemgeo.2022.120724
973 Corentin, P., Pucéat, E., Pellenard, P., Guiraud, M., Blondet, J., Bayon, G., Adatte, T., 2024.
974 Enhanced erosion and silicate weathering of the West African craton during the late
975 Cretaceous cooling evidenced by mineralogical and HfNd isotope proxies. Mar. Geol.
976 476, 107374. https://doi.org/10.1016/j.margeo.2024.107374
977 Corentin, P., Pucéat, E., Pellenard, P., Guiraud, M., Blondet, J., Bayon, G., Adatte, T., 2023.
978 Late Cretaceous evolution of chemical weathering at the northeastern South American
979 margin inferred from mineralogy and Hf-Nd isotopes. Mar. Geol. 455, 106968.
980 https://doi.org/10.1016/j.margeo.2022.106968

Cullers, R.L., Arnold, B., Lee, M., Wolf, C.W., 1975. Rare earth distintions in clay minerals and in the clay-sized fraction of the Lower Permian Havensville and Eskridge shales of Kansas and Oklahoma. Geochimica et Cosmochimica Acta 39, 1691–1703.

de Vera, J., Granado, P., McClay, K., 2010. Structural evolution of the Orange Basin gravity-driven system, offshore Namibia. Mar. Pet. Geol. 27, 223–237. https://doi.org/10.1016/j.marpetgeo.2009.02.003

De Wit, M.C.J., Ward, J.D., Bamford, M.K., Roberts, M.J., 2009. THE SIGNIFICANCE OF THE CRETACEOUS DIAMONDIFEROUS GRAVEL DEPOSIT AT MAHURA MUTHLA, NORTHERN CAPE PROVINCE, SOUTH AFRICA. South Afr. J. Geol. 112, 89–108. https://doi.org/10.2113/gssajg.112.2.89

De Wit, MichielC.J., 1999. The distribution and stratigraphy of inland alluvial diamond deposits in South Africa. Afr. Geosci. Rev. Spec. Ed. 3, 19–33.

Dera, G., Pellenard, P., Neige, P., Deconinck, J.-F., Pucéat, E., Dommergues, J.-L., 2009. Distribution of clay minerals in Early Jurassic Peritethyan seas: Palaeoclimatic significance inferred from multiproxy comparisons. Palaeogeogr. Palaeoclimatol. Palaeoecol. 271, 39–51. https://doi.org/10.1016/j.palaeo.2008.09.010

Dia, A., Allègre, C.J., Erlank, A.J., 1990. The development of continental crust through geological time: the South African case. Earth Planet. Sci. Lett. 98, 74–89. https://doi.org/10.1016/0012-821X(90)90089-G

Dinis, P.A., Garzanti, E., Hahn, A., Vermeesch, P., Cabral-Pinto, M., 2020. Weathering indices as climate proxies. A step forward based on Congo and SW African river muds. Earth-Sci. Rev. 201, 103039. https://doi.org/10.1016/j.earscirev.2019.103039

Dudus, V., Dizon, E., Doczy, G., Meikle, G., Heath, R., Kumar, D., Cleaver, R., Barrett, A., McLellen, A., Peel, A., Gleghorn, I., 1992. Final well report O-A1. SOEKOR, South Africa.

1006 Dymond, J., Suess, E., Lyle, M., 1992. Barium in Deep-Sea Sediment: A Geochemical Proxy
1007 for Paleoproductivity. Paleoceanography 7, 163–181.
1008 https://doi.org/10.1029/92PA00181
1009 Eagle, M., Paytan, A., Arrigo, K.R., van Dijken, G., Murray, R.W., 2003. A comparison
1010 between excess barium and barite as indicators of carbon export. Paleoceanography 18.
1011 https://doi.org/10.1029/2002PA000793
1012 Fagel, N., André, L., Chamley, H., Debrabant, P., Jolivet, L., 1992. Clay sedimentation in the
1013 Japan Sea since the Early Miocene: influence of source-rock and hydrothermal activity.
1014 Sediment. Geol. 80, 27–40. https://doi.org/10.1016/0037-0738(92)90029-Q
1015 Fagel, N., Robert, C., Preda, M., Thorez, J., 2001. Smectite composition as a tracer of deep
1016 circulation: the case of the Northern North Atlantic. Mar. Geol. 172, 309–330.
1017 https://doi.org/10.1016/S0025-3227(00)00123-7
1018 Flowers, R.M., Schoene, B., 2010. (U-Th)/He thermochronometry constraints on unroofing of
1019 the eastern Kaapvaal craton and significance for uplift of the southern African Plateau.
1020 Geology 38, 827–830. https://doi.org/10.1130/G30980.1
1021 Freslon, N., Bayon, G., Birot, D., Bollinger, C., Barrat, J.A., 2011. Determination of rare earth
1022 elements and other trace elements (Y, Mn, Co, Cr) in seawater using Tm addition and
1023 Mg(OH)2 co-precipitation. Talanta 85, 582–587.
1024 https://doi.org/10.1016/j.talanta.2011.04.023
1025 Friedrich, O., Norris, R.D., Erbacher, J., 2012. Evolution of middle to Late Cretaceous
1026 oceans—A 55 m.y. record of Earth's temperature and carbon cycle. Geology 40, 107–
1027 110. https://doi.org/10.1130/G32701.1
1028 Gabet, E., Mudd, S., 2009. A theoretical model coupling chemical weathering rate with
1029 denudation rates. Geology 37, 151–154. https://doi.org/10.1130/G25270A.1

1030 Gaillardet, J., Dupré, B., Louvat, P., Allègre, C.J., 1999. Global silicate weathering and CO2
1031 consumption rates deduced from the chemistry of large rivers. Chem. Geol. 159, 3–30.
1032 https://doi.org/10.1016/S0009-2541(99)00031-5
1033 Gaitan, C.E., Pucéat, E., Pellenard, P., Blondet, J., Bayon, G., Adatte, T., Israel, C., Robin, C.,
1034 Guillocheau, F., 2023. Late Cretaceous erosion and chemical weathering record in the
1035 offshore Cape Basin: Source-to-sink system from Hf Nd isotopes and clay mineralogy.
1036 Mar. Geol. 466, 107187. https://doi.org/10.1016/j.margeo.2023.107187
1037 Gallagher, K., Brown, R., 1999. Denudation and uplift at passive margins: the record on the
1038 Atlantic Margin of southern Africa. Philos. Trans. R. Soc. Lond. Ser. Math. Phys. Eng.
1039 Sci. 357, 835–859. https://doi.org/10.1098/rsta.1999.0354
1040 Garçon, M., Chauvel, C., France-Lanord, C., Huyghe, P., Lavé, J., 2013. Continental
1041 sedimentary processes decouple Nd and Hf isotopes. Geochim. Cosmochim. Acta 121,
1042 177–195. https://doi.org/10.1016/j.gca.2013.07.027
1043 Gibbs, R.J., 1977. Clay mineral segregation in the marine environment. J. Sediment. Res. 47,
1044 237–243. https://doi.org/10.1306/212F713A-2B24-11D7-8648000102C1865D
1045 Gladczenko, T.P., Hinz, K., Eldholm, O., Meyer, H., Neben, S., Skogseid, J., 1997. South
1046 Atlantic volcanic margins. J. Geol. Soc. 154, 465–470.
1047 https://doi.org/10.1144/gsjgs.154.3.0465
1048 Goldstein, S.J., Jacobsen, S.B., 1988. Nd and Sr isotopic systematics of river water suspended
1049 material: implications for crustal evolution. Earth Planet. Sci. Lett. 87, 249–265.
1050 https://doi.org/10.1016/0012-821X(88)90013-1
1051 Goldstein, S.L., O'Nions, R.K., Hamilton, P.J., 1984. A Sm-Nd isotopic study of atmospheric
1052 dusts and particulates from major river systems. Earth Planet. Sci. Lett. 70, 221–236.
1053 https://doi.org/10.1016/0012-821X(84)90007-4

1054 Gomes, A.S., Vasconcelos, P.M., 2021. Geochronology of the Paraná-Etendeka large igneous
1055 province. Earth-Sci. Rev. 220, 103716.
1056 https://doi.org/10.1016/j.earscirev.2021.103716
1057 Goudie, A.S., 2005. The drainage of Africa since the Cretaceous. Geomorphology 67, 437–
1058 456. https://doi.org/10.1016/j.geomorph.2004.11.008
1059 Green, P.F., Duddy, I.R., Japsen, P., Bonow, J.M., Malan, J.A., 2017. Post-breakup burial and
1060 exhumation of the southern margin of Africa. Basin Res. 29, 96–127.
1061 https://doi.org/10.1111/bre.12167
1062 Gutjahr, M., Frank, M., Stirling, C.H., Klemm, V., van de Flierdt, T., Halliday, A.N., 2007.
1063 Reliable extraction of a deepwater trace metal isotope signal from Fe–Mn oxyhydroxide
1064 coatings of marine sediments. Chem. Geol. 242, 351–370.
1065 https://doi.org/10.1016/j.chemgeo.2007.03.021
1066 Hawkesworth, C.J., Marsh, J.S., Duncan, A.R., Erlank, A.J., Norry, M.J., 1984. The role of
1067 continental lithosphere in the generation of the Karoo volcanic rocks: evidence from
1068 combined Nd-and Sr-isotope studies.
1069 Hilton, R.G., West, A.J., 2020. Mountains, erosion and the carbon cycle. Nat. Rev. Earth
1070 Environ. 1, 284–299. https://doi.org/10.1038/s43017-020-0058-6
1071 Hindshaw, R.S., Tosca, N.J., Piotrowski, A.M., Tipper, E.T., 2018. Clay mineralogy, strontium
1072 and neodymium isotope ratios in the sediments of two High Arctic catchments
1073 (Svalbard). Earth Surf. Dyn. 6, 141–161. https://doi.org/10.5194/esurf-6-141-2018
1074 Hirsch, K.K., Scheck-Wenderoth, M., van Wees, J.-D., Kuhlmann, G., Paton, D.A., 2010.
1075 Tectonic subsidence history and thermal evolution of the Orange Basin. Mar. Pet. Geol.
1076 27, 565–584. https://doi.org/10.1016/j.marpetgeo.2009.06.009

1077 Inoue, A., Utada, M., 1991. Smectite-to-chlorite transformation in thermally metamorphosed
1078 volcanoclastic rocks in the Kamikita area, northern Honshu, Japan. Am. Mineral. 76,
1079 628–640.
1080 Jochum, K.P., Weis, U., Schwager, B., Stoll, B., Wilson, S.A., Haug, G.H., Andreae, M.O.,
1081 Enzweiler, J., 2016. Reference Values Following ISO Guidelines for Frequently
1082 Requested Rock Reference Materials. Geostand. Geoanalytical Res. 40, 333–350.
1083 https://doi.org/10.1111/j.1751-908X.2015.00392.x
1084 John, C.M., Banerjee, N.R., Longstaffe, F.J., Sica, C., Law, K.R., Zachos, J.C., 2012. Clay
1085 assemblage and oxygen isotopic constraints on the weathering response to the
1086 Paleocene-Eocene thermal maximum, east coast of North America. Geology 40, 591–
1087 594. https://doi.org/10.1130/G32785.1
1088 Jourdan, F., Bertrand, H., Schärer, U., Blichert-Toft, J., Féraud, G., Kampunzu, A.B., 2007.
1089 Major and Trace Element and Sr, Nd, Hf, and Pb Isotope Compositions of the Karoo
1090 Large Igneous Province, Botswana–Zimbabwe: Lithosphere vs Mantle Plume
1091 Contribution. J. Petrol. 48, 1043–1077. https://doi.org/10.1093/petrology/egm010
1092 Jungslager, E.H.A., 1999. Petroleum habitats of the Atlantic margin of South Africa. Geol.
1093 Soc. Lond. Spec. Publ. 153, 153–168. https://doi.org/10.1144/GSL.SP.1999.153.01.10
1094 Ketzer, M., Morad, S., Amorosi, A., 2003. Predictive Diagenetic Clay-Mineral Distribution in
1095 Siliciclastic Rocks within a Sequence Stratigraphic Framework, in: International
1096 Association of Sedimentologists Special Publications. pp. 43–61.
1097 https://doi.org/10.1002/9781444304336.ch2
1098 Koopmann, H., Schreckenberger, B., Franke, D., Becker, K., Schnabel, M., 2016. The late
1099 rifting phase and continental break-up of the southern South Atlantic: the mode and
1100 timing of volcanic rifting and formation of earliest oceanic crust. Geol. Soc. Lond.
1101 Spec. Publ. 420, 315–340. https://doi.org/10.1144/SP420.2

1102 Kounov, A., Viola, G., de Wit, M., Andreoli, M.A.G., 2009. Denudation along the Atlantic
1103 passive margin: new insights from apatite fission-track analysis on the western coast of
1104 South Africa. Geol. Soc. Lond. Spec. Publ. 324, 287–306.
1105 https://doi.org/10.1144/SP324.19
1106 Kounov, A., Viola, G., Dunkl, I., Frimmel, H.E., 2013. Southern African perspectives on the
1107 long-term morpho-tectonic evolution of cratonic interiors. Tectonophysics 601, 177–
1108 191. https://doi.org/10.1016/j.tecto.2013.05.009
1109 Lanson, B., Beaufort, D., Berger, G., Bauer, A., Cassagnabère, A., Meunier, A., 2002.
1110 Authigenic kaolin and illitic minerals during burial diagenesis of sandstones: a review.
1111 Clay Miner. 37, 1–22. https://doi.org/10.1180/0009855023710014
1112 Lanson, B., Sakharov, B., Claret, F., Drits, V., 2009. Diagenetic Smectite-to-Illite Transition
1113 in Clay-Rich Sediments: A Reappraisal of X-Ray Diffraction Results Using the Multi-
1114 Specimen Method. Am. J. Sci. 309, 476–516. https://doi.org/10.2475/06.2009.03
1115 Lidman, F., Laudon, H., Taberman, I., Köhler, S., 2019. Eu anomalies in soils and soil water
1116 from a boreal hillslope transect – A tracer for Holocene lanthanide transport? Geochim.
1117 Cosmochim. Acta 267, 147–163. https://doi.org/10.1016/j.gca.2019.09.014
1118 Liu, K., Feng, Q., Shen, J., Khan, M., Planavsky, N.J., 2018. Increased productivity as a
1119 primary driver of marine anoxia in the Lower Cambrian. Palaeogeogr. Palaeoclimatol.
1120 Palaeoecol. 491, 1–9. https://doi.org/10.1016/j.palaeo.2017.11.007
1121 Lugmair, G.W., Marti, K., 1977. SmNdPu timepieces in the Angra dos Reis meteorite. Earth
1122 Planet. Sci. Lett. 35, 273–284. https://doi.org/10.1016/0012-821X(77)90131-5
1123 Maher, K., 2010. The dependence of chemical weathering rates on fluid residence time. Earth
1124 Planet. Sci. Lett. 294, 101–110. https://doi.org/10.1016/j.epsl.2010.03.010

1125 Maher, K., Chamberlain, C.P., 2014. Hydrologic Regulation of Chemical Weathering and the
1126 Geologic Carbon Cycle. Science 343, 1502–1504.
1127 https://doi.org/10.1126/science.1250770
1128 McCulloch, M.T., Wasserburg, G.J., 1978. Sm-Nd and Rb-Sr Chronology of Continental Crust
1129 Formation: Times of addition to continents of chemically fractionated mantle-derived
1130 materials are determined. Science 200, 1003–1011.
1131 https://doi.org/10.1126/science.200.4345.1003
1132 Moore, D.M., Reynolds, J., 1997. X-ray Diffraction and the Identification and Analysis of Clay
1133 Minerals, Second edition. ed. Oxford University Press, London.
1134 Moulin, M., Aslanian, D., Unternehr, P., 2010. A new starting point for the South and
1135 Equatorial Atlantic Ocean. Earth-Sci. Rev. 98, 1–37.
1136 https://doi.org/10.1016/j.earscirev.2009.08.001
1137 Murphy, D.P., Thomas, D.J., 2013. The evolution of Late Cretaceous deep-ocean circulation
1138 in the Atlantic basins: Neodymium isotope evidence from South Atlantic drill sites for
1139 tectonic controls. Geochem. Geophys. Geosystems 14, 5323–5340.
1140 https://doi.org/10.1002/2013GC004889
1141 Nadeau, P.H., Wilson, M.J., McHardy, W.J., Tait, J.M., 1985. The conversion of smectite to
1142 illite during diagenesis: evidence from some illitic clays from bentonites and
1143 sandstones. Mineral. Mag. 49, 393–400.
1144 https://doi.org/10.1180/minmag.1985.049.352.10
1145 NASA JPL, 2013. NASA Shuttle Radar Topography Mission Global 30 arc second.
1146 https://doi.org/10.5067/MEASURES/SRTM/SRTMGL30.002
1147 Natland, J.H., 1978. COMPOSITION, PROVENANCE, AND DIAGENESIS OF
1148 CRETACEOUS CLASTIC SEDIMENTS DRILLED ON THE ATLANTIC

CONTINENTAL RISE OFF SOUTHERN AFRICA, DSDP SITE 361. IMPLICATIONS FOR THE EARLY CIRCULATION OF THE SOUTH ATLANTIC.

Nesbitt, H.W., Young, G.M., 1982. Early Proterozoic climates and plate motions inferred from major element chemistry of lutites. Nature 299, 715–717. https://doi.org/10.1038/299715a0

Nozaki, Y., 2001. Rare Earth Elements and their Isotopes in the Ocean, in: Steele, J.H. (Ed.), Encyclopedia of Ocean Sciences. Academic Press, Oxford, pp. 2354–2366. https://doi.org/10.1006/rwos.2001.0284

O'Brien, C.L., Robinson, S.A., Pancost, R.D., Sinninghe Damsté, J.S., Schouten, S., Lunt, D.J., Alsenz, H., Bornemann, A., Bottini, C., Brassell, S.C., Farnsworth, A., Forster, A., Huber, B.T., Inglis, G.N., Jenkyns, H.C., Linnert, C., Littler, K., Markwick, P., McAnena, A., Mutterlose, J., Naafs, B.D.A., Püttmann, W., Sluijs, A., van Helmond, N.A.G.M., Vellekoop, J., Wagner, T., Wrobel, N.E., 2017. Cretaceous sea-surface temperature evolution: Constraints from TEX86 and planktonic foraminiferal oxygen isotopes. Earth-Sci. Rev. 172, 224–247. https://doi.org/10.1016/j.earscirev.2017.07.012

Patchett, P.J., White, W.M., Feldmann, H., Kielinczuk, S., Hofmann, A.W., 1984. Hafnium/rare earth element fractionation in the sedimentary system and crustal recycling into the Earth's mantle. Earth Planet. Sci. Lett. 69, 365–378. https://doi.org/10.1016/0012-821X(84)90195-X

Paton, D.A., van der Spuy, D., di Primio, R., Horsfield, B., 2008. Tectonically induced adjustment of passive-margin accommodation space; influence on the hydrocarbon potential of the Orange Basin, South Africa. AAPG Bull. 92, 589–609. https://doi.org/10.1306/12280707023

1173 Paton, Douglas.A., di Primio, R., Kuhlmann, G., van der Spuy, D., Horsfield, B., 2007. Insights
1174 into the Petroleum System Evolution of the southern Orange Basin, South Africa. South
1175 Afr. J. Geol. 110, 261–274. https://doi.org/10.2113/gssajg.110.2-3.261
1176 Paytan, A., Griffith, E.M., 2007. Marine barite: Recorder of variations in ocean export
1177 productivity. Deep Sea Res. Part II Top. Stud. Oceanogr., The Role of Marine Organic
1178 Carbon and Calcite Fluxes in Driving Global Climate Change, Past and Future 54, 687–
1179 705. https://doi.org/10.1016/j.dsr2.2007.01.007
1180 Pearson, M.J., Small, J.S., 1988. Illite-smectite diagenesis and palaeotemperatures in Northern
1181 North Sea Quaternary to Mesozoic shale sequences. Clay Miner. 23, 109–132.
1182 https://doi.org/10.1180/claymin.1988.023.2.01
1183 Petschick, R., 2001. MacDiff Ver. 4.2. 5.
1184 Petschick, R., Kuhn, G., Gingele, F., 1996. Clay mineral distribution in surface sediments of
1185 the South Atlantic: sources, transport, and relation to oceanography. Mar. Geol. 130,
1186 203–229. https://doi.org/10.1016/0025-3227(95)00148-4
1187 Peucker-Ehrenbrink, B., Miller, M.W., Arsouze, T., Jeandel, C., 2010. Continental bedrock
1188 and riverine fluxes of strontium and neodymium isotopes to the oceans. Geochem.
1189 Geophys. Geosystems 11. https://doi.org/10.1029/2009GC002869
1190 Pogge von Strandmann, P.A.E., Jenkyns, H.C., Woodfine, R.G., 2013. Lithium isotope
1191 evidence for enhanced weathering during Oceanic Anoxic Event 2. Nat. Geosci. 6, 668–
1192 672. https://doi.org/10.1038/ngeo1875
1193 Raab, M.J., Brown, R.W., Gallagher, K., Carter, A., Weber, K., 2002. Late Cretaceous
1194 reactivation of major crustal shear zones in northern Namibia: constraints from apatite
1195 fission track analysis. Tectonophysics 349, 75–92. https://doi.org/10.1016/S0040-
1196 1951(02)00047-1

1197 Rabinowitz, P.D., LaBrecque, J., 1979. The Mesozoic South Atlantic Ocean and evolution of
1198 its continental margins. J. Geophys. Res. Solid Earth 84, 5973–6002.
1199 https://doi.org/10.1029/JB084iB11p05973
1200 Rayner, R.J., Bamford, M.K., Brothers, D.J., Dippenaar-Schoeman, A.S., McKay, I.J.,
1201 Oberprieler, R.G., Watersl, S.B., 1997. CRETACEOUS FOSSILS FROM THE
1202 ORAPA DIAMOND MINE 11.
1203 Reid, D.L., 1997. Sm-Nd age and REE geochemistry of Proterozoic arc-related igneous rocks
1204 in the Richtersveld subprovince, Namaqua Mobile Belt, Southern Africa. J. Afr. Earth
1205 Sci. 24, 621–633. https://doi.org/10.1016/S0899-5362(97)00084-5
1206 Renne, P.R., 2015. Age and duration of the Paraná-Etendeka flood basalts and related plumbing
1207 system, in: AGU Fall Meeting Abstracts. pp. T32D-06.
1208 Riebe, C.S., Kirchner, J.W., Finkel, R.C., 2004. Erosional and climatic effects on long-term
1209 chemical weathering rates in granitic landscapes spanning diverse climate regimes.
1210 Earth Planet. Sci. Lett. 224, 547–562. https://doi.org/10.1016/j.epsl.2004.05.019
1211 Sandersen, A., 2006. A palynological investigation of the offshore Cretaceous sequence on the
1212 south-west coast of South Africa (PhD Thesis). Uni- versity of the Witswatersrand.
1213 Schoepfer, S.D., Shen, J., Wei, H., Tyson, R.V., Ingall, E., Algeo, T.J., 2015. Total organic
1214 carbon, organic phosphorus, and biogenic barium fluxes as proxies for paleomarine
1215 productivity. Earth-Sci. Rev., Global review of the Permian-Triassic mass extinction
1216 and subsequent recovery: Part II 149, 23–52.
1217 https://doi.org/10.1016/j.earscirev.2014.08.017
1218 Séranne, M., Anka, Z., 2005. South Atlantic continental margins of Africa: A comparison of
1219 the tectonic vs climate interplay on the evolution of equatorial west Africa and SW
1220 Africa margins. J. Afr. Earth Sci. 43, 283–300.
1221 https://doi.org/10.1016/j.jafrearsci.2005.07.010

1222 Smith, R.M.H., 1986. Sedimentation and palaeoenvironments of Late Cretaceous crater-lake
1223 deposits in Bushmanland, South Africa. Sedimentology 33, 369–386.
1224 https://doi.org/10.1111/j.1365-3091.1986.tb00542.x
1225 Söderlund, U., Patchett, P.J., Vervoort, J.D., Isachsen, C.E., 2004. The 176Lu decay constant
1226 determined by Lu–Hf and U–Pb isotope systematics of Precambrian mafic intrusions.
1227 Earth Planet. Sci. Lett. 219, 311–324. https://doi.org/10.1016/S0012-821X(04)00012-
1228 3
1229 Stanley, J.R., Braun, J., Baby, G., Guillocheau, F., Robin, C., Flowers, R.M., Brown, R.,
1230 Wildman, M., Beucher, R., 2021. Constraining Plateau Uplift in Southern Africa by
1231 Combining Thermochronology, Sediment Flux, Topography, and Landscape Evolution
1232 Modeling. J. Geophys. Res. Solid Earth 126. https://doi.org/10.1029/2020JB021243
1233 Stanley, J.R., Flowers, R.M., 2020. Mesozoic denudation history of the lower Orange River
1234 and eastward migration of erosion across the southern African Plateau. Lithosphere 12,
1235 74–87. https://doi.org/10.1130/L1121.1
1236 Stanley, J.R., Flowers, R.M., Bell, D.R., 2015. Erosion patterns and mantle sources of
1237 topographic change across the southern African Plateau derived from the shallow and
1238 deep records of kimberlites. Geochem. Geophys. Geosystems 16, 3235–3256.
1239 https://doi.org/10.1002/2015GC005969
1240 Stanley, J.R., Flowers, R.M., Bell, D.R., 2013. Kimberlite (U-Th)/He dating links surface
1241 erosion with lithospheric heating, thinning, and metasomatism in the southern African
1242 Plateau. Geology 41, 1243–1246. https://doi.org/10.1130/G34797.1
1243 Stevenson, I.R., Mcmillan, I.K., 2004. Incised valley fill stratigraphy of the Upper Cretaceous
1244 succession, proximal Orange basin, Atlantic margin of southern Africa. J. Geol. Soc.
1245 Lond. 185–208.

1246 Surdam, R.C., Crossey, L.J., 1987. Integrated Diagenetic Modeling: A Process-Oriented
1247 Approach for Clastic Systems. Annu. Rev. Earth Planet. Sci. 15, 141–170.
1248 https://doi.org/10.1146/annurev.ea.15.050187.001041
1249 Tanaka, T., Togashi, S., Kamioka, H., Amakawa, H., Kagami, H., Hamamoto, T., Yuhara, M.,
1250 Orihashi, Y., Yoneda, S., Shimizu, H., Kunimaru, T., Takahashi, K., Yanagi, T.,
1251 Nakano, T., Fujimaki, H., Shinjo, R., Asahara, Y., Tanimizu, M., Dragusanu, C., 2000.
1252 JNdi-1: a neodymium isotopic reference in consistency with LaJolla neodymium.
1253 Chem. Geol. 168, 279–281. https://doi.org/10.1016/S0009-2541(00)00198-4
1254 Taylor, S.R., McLennan, S.M., 1985. The continental crust: Its composition and evolution.
1255 Tinker, J., de Wit, M., Brown, R., 2008. Mesozoic exhumation of the southern Cape, South
1256 Africa, quantified using apatite fission track thermochronology. Tectonophysics 455,
1257 77–93. https://doi.org/10.1016/j.tecto.2007.10.009
1258 Torsvik, T.H., Rousse, S., Labails, C., Smethurst, M.A., 2009. A new scheme for the opening
1259 of the South Atlantic Ocean and the dissection of an Aptian salt basin. Geophys. J. Int.
1260 177, 1315–1333. https://doi.org/10.1111/j.1365-246X.2009.04137.x
1261 Valicenti, V.H., Pringle, A.A., Benson, J.M., 1993. Biostratigraphic report on borehole O-A1.
1262 SOEKOR.
1263 Van Der Spuy, D., 2003. Aptian source rocks in some South African Cretaceous basins. Geol.
1264 Soc. Lond. Spec. Publ. 207, 185–202. https://doi.org/10.1144/GSL.SP.2003.207.10
1265 Velde, B., 1995. Origin and mineralogy of clays: clays and the environment. Springer Science
1266 & Business Media.
1267 Vervoort, J.D., Plank, T., Prytulak, J., 2011. The Hf–Nd isotopic composition of marine
1268 sediments. Geochim. Cosmochim. Acta 75, 5903–5926.
1269 https://doi.org/10.1016/j.gca.2011.07.046

Warr, L.N., 2022. Earth's clay mineral inventory and its climate interaction: A quantitative assessment. Earth-Sci. Rev. 234, 104198. https://doi.org/10.1016/j.earscirev.2022.104198

Weis, D., Kieffer, B., Maerschalk, C., Pretorius, W., Barling, J., 2005. High-precision Pb-Sr-Nd-Hf isotopic characterization of USGS BHVO-1 and BHVO-2 reference materials: Pb-Sr-Nd-Hf CHARACTERIZATION. Geochem. Geophys. Geosystems 6. https://doi.org/10.1029/2004GC000852

West, A., Galy, A., Bickle, M., 2005. Tectonic and climatic controls on silicate weathering. Earth Planet. Sci. Lett. 235, 211–228. https://doi.org/10.1016/j.epsl.2005.03.020

Wildman, M., Brown, R., Beucher, R., Persano, C., Stuart, F., Gallagher, K., Schwanethal, J., Carter, A., 2016. The chronology and tectonic style of landscape evolution along the elevated Atlantic continental margin of South Africa resolved by joint apatite fission track and (U-Th-Sm)/He thermochronology. Tectonics 35, 511–545. https://doi.org/10.1002/2015TC004042

Wildman, M., Brown, R., Watkins, R., Carter, A., Gleadow, A., Summerfield, M., 2015. Post break-up tectonic inversion across the southwestern cape of South Africa: New insights from apatite and zircon fission track thermochronometry. Tectonophysics 654, 30–55. https://doi.org/10.1016/j.tecto.2015.04.012

Wildman, M., Gallagher, K., Chew, D., Carter, A., 2021. From sink to source: Using offshore thermochronometric data to extract onshore erosion signals in Namibia. Basin Res. 33, 1580–1602. https://doi.org/10.1111/bre.12527

Yobo, L., Brandon, A.D., Holmden, C., Lau, K.V., Eldrett, J., 2021. Changing inputs of continental and submarine weathering sources of Sr to the oceans during OAE 2. Geochim. Cosmochim. Acta 303, 205–222. https://doi.org/10.1016/j.gca.2021.03.013

Zhao, Y., Wei, W., Li, S., Yang, T., Zhang, R., Somerville, I., Santosh, M., Wei, H., Wu, J., Yang, J., Chen, W., Tang, Z., 2021. Rare earth element geochemistry of carbonates as a proxy for deep-time environmental reconstruction. Palaeogeogr. Palaeoclimatol. Palaeoecol. 574, 110443. https://doi.org/10.1016/j.palaeo.2021.110443

Zhu, Y., Itoh, A., 2021. Pseudo isotope dilution (PID) as an approach for correcting barium-related spectral interferences on the measurement of europium by inductively coupled plasma mass spectrometry (ICP-MS). Anal. Chim. Acta 1180, 338854. https://doi.org/10.1016/j.aca.2021.338854

Data availability

All the data are provided in the supplementary materials attached to this paper.

Acknowledgments

We thank the South African Petroleum Agency (SAPA) for providing access to the material (borehole O-A1). This project has received funding from the European Union's Horizon 2020 research and innovation program under the Marie Sklodowska-Curie grant agreement No 860383 and support from the project ANR RISE. We thank Rémi Chassagnon (Laboratory ICB, University of Burgundy) and INRAe of Dijon, Ludovic Bruneau of the Biogéosciences Laboratory and GISMO analytical Platform (University of Burgundy), Marie-Laure Rouget and Bleuenn Gueguen of the Pôle Spectrométrie Océan (Brest), Olivier Reubi of the Institut des Sciences de la Terre (ISTE-Lausanne), and Philippe Telouk of the ENS Lyon for their analytical support.