CHAPTER 4.4

CHEMOSTRATIGRAPHY☆

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Contents

4.4.1. INTRODUCTION

Isotopic chemostratigraphy was first applied to Neoproterozoic successions in the RPC by Boggiani (1998) and Kawashita et al. (1999a,b), and a compilation of the data was published by Gaucher (2000). A relatively large database is now available for the Mina Verdún, Arroyo del Soldado and Sierras Bayas groups (Gaucher et al., 2003, 2004c,d, 2006, 2007a,c; Gómez Peral et al., 2007), as well as preliminary data for the upper Piedras de Afilar Formation (Pamoukaghlian et al., 2006).

4.4.2. CARBON-ISOTOPE CHEMOSTRATIGRAPHY

4.4.2.1. Mina Verdún Group

A total of 88 carbon- and oxygen-isotope analyses on carbonates of the Mina Verdún Group were presented by Gaucher et al. (2006, 2007a). Three sections were studied, namely: (1) the stratotype of the group at Mina Verdún, (2) Burgueño Quarry and (3) Paso del Molino (Figure 4.2.2). δ^{13}C versus δ^{18}O crossplots display strong co-variation of both parameters and more negative values for the latter two sections, showing that C-isotope composition has been affected by post-depositional processes (Figure 4.4.1). However, corresponding Mn/Sr values are 0.3 for limestones at the Paso del Molino section, suggesting that the isotopic ratios are near-primary (Jacobsen and Kaufman, 1999). The strong decolouration of limestones at Burgueño Quarry and Paso del Molino, compared to the same levels at the stratotype, shows that organic carbon was oxidated, leading to production of CO_2 enriched in ^{12}C. This was later incorporated into the carbonates during early diagenesis,

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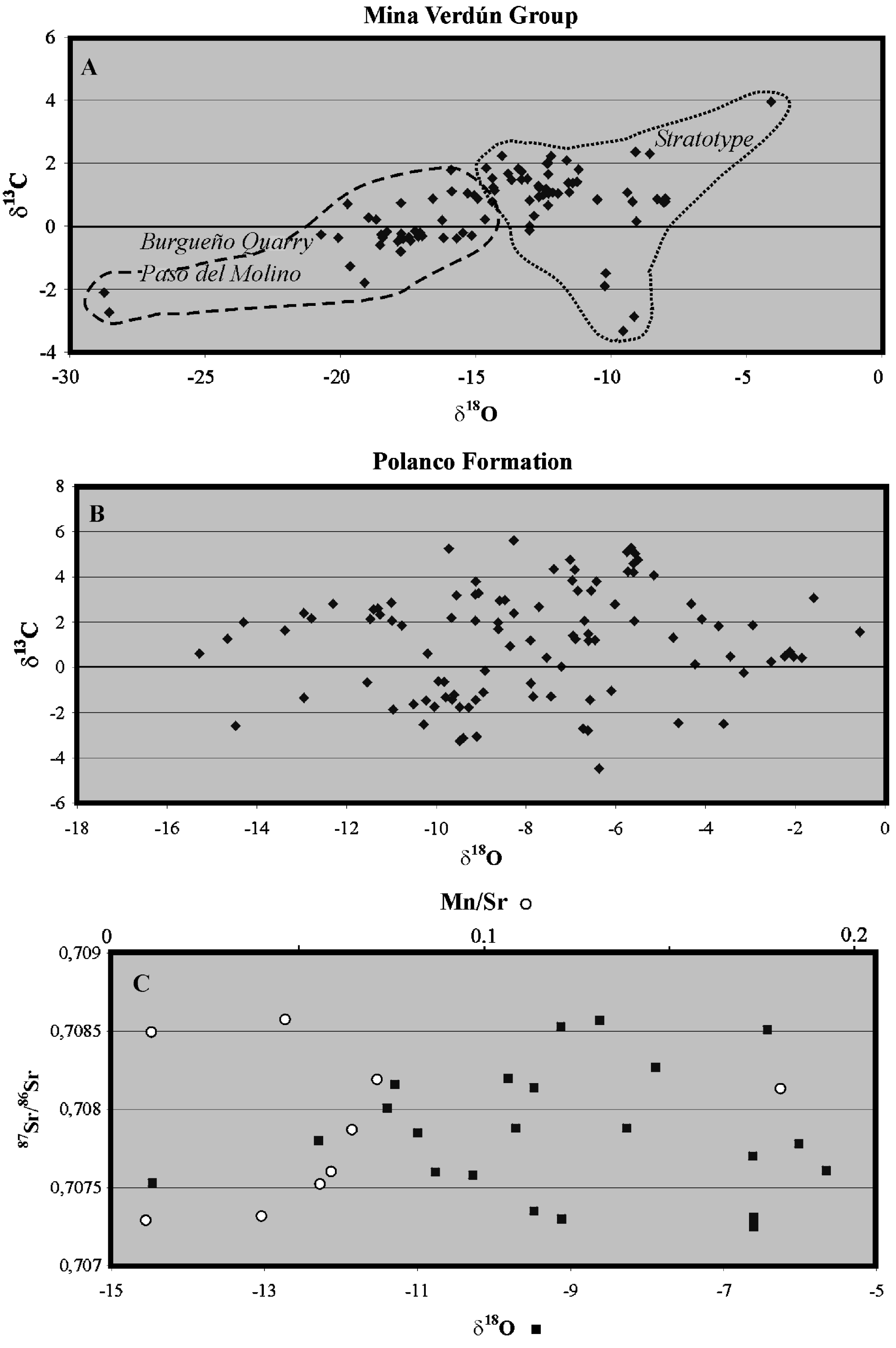

Figure 4.4.1 (A) $\delta^{13}C$ versus $\delta^{18}O$ crossplot for 88 samples of the Mina Verdún Group. Note altered nature (linear array) of samples from the Burgueño Quarry and Paso del Molino sections, contrasting with the primary isotopic signature at the stratotype. (B) $\delta^{13}C$ versus $\delta^{18}O$ crossplot of more than 100 samples from the Polanco Formation throughout the basin. Note lack of correlation between both parameters. (C) $^{87}Sr/^{86}Sr$ versus $\delta^{18}O$ (black squares) and $^{87}Sr/^{86}Sr$ versus Mn/Sr (white circles) for limestones of the Polanco Formation (22 samples). Lack of co-variance strongly suggests that $^{87}Sr/^{86}Sr$ values reflect seawater composition.

resulting in lower $\delta^{13}C$ ratios (Murata et al., 1969; De Giovanni et al., 1974). These reactions do not necessarily involve Sr or Mn remobilisation, thus explaining the low Mn/Sr ratios.

At the stratotype in Mina Verdún, no co-variation of $\delta^{13}C$ versus $\delta^{18}O$ is observed (Figure 4.4.1), and a very consistent trend from negative to positive values is recorded (Gaucher et al., 2006). $\delta^{13}C$ is higher by 3‰ PDB than at corresponding stratigraphic levels in the other sections, and organic matter is well preserved. Therefore, we

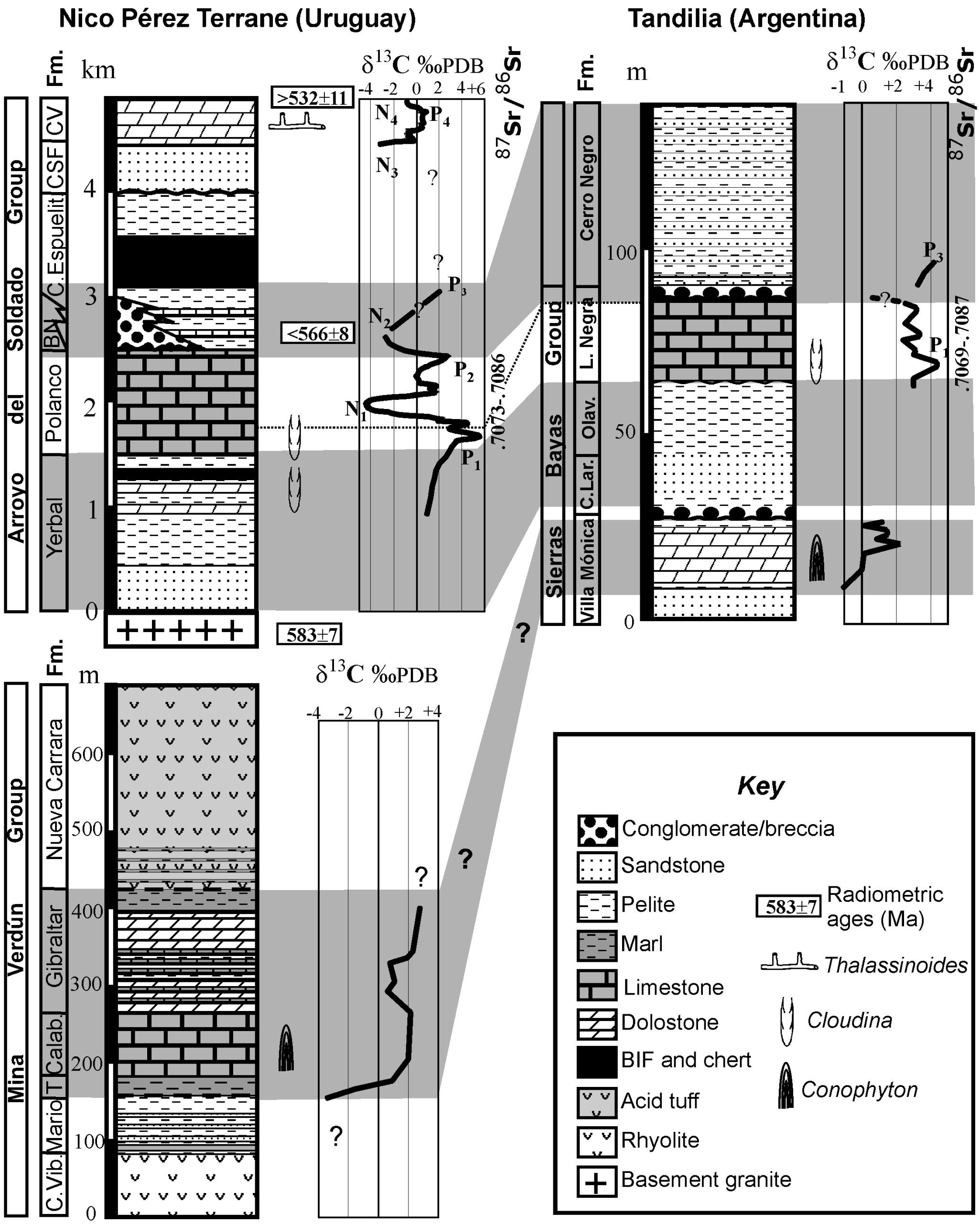

Figure 4.4.2 Available chemostratigraphic data for the Arroyo del Soldado, Mina Verdún and Sierras Bayas groups and inferred correlations. Arroyo del Soldado Group — BN: Barriga Negra Formation; C.Espuelit.: Cerro Espuelitas Formation; CSF: Cerros San Francisco Formation; CV: Cerro Victoria Formation. Radiometric ages shown are 583 ± 7 Ma for the Mangacha Granite (Gaucher et al., 2008b), 566 ± 8 Ma for the youngest detrital zircon in the Barriga Negra Formation (Blanco et al., 2009) and 532 ± 11 Ma for the Guazunambí Granite, intrusive in the ASG (Kawashita et al., 1999a). Sierras Bayas Group — C.Lar.: Cerro Largo Formation; Olav.: Olavarría Formation; L.Negra: Loma Negra Formation. Chemostratigraphic data from Kawashita et al. (1999b) and Gómez Peral et al. (2007). Mina Verdún Group (Gaucher et al., 2007a) — C.Vib.: Cerro de las Víboras Formation; Mario: Don Mario Formation; T: La Toma Formation; Calab.: El Calabozo Formation. Modified from Gaucher et al. (2005b).

conclude that carbon-isotopic ratios at Mina Verdún reflect coeval seawater composition. The $\delta^{13}C$ curve thus obtained (Figure 4.4.2) shows a negative excursion at the base (La Toma Formation) to −3.3‰ PDB, steadily rising to values around 1.5‰ PDB up section. For 240 m of section (El Calabozo and Gibraltar formations) only positive $\delta^{13}C$ values occur, varying between 0.15‰ and 4.0‰ PDB, but mostly around a plateau centred at 2‰ PDB (Gaucher et al., 2006). This plateau is the most prominent chemostratigraphic feature of the Mina Verdún Group (Figure 4.4.2).

The $\delta^{13}C$ curve obtained has the following salient features: (a) moderate amplitude of $\delta^{13}C$ excursions of up to 5.5‰ (transition between La Toma and El Calabozo formations), (b) $\delta^{13}C$ values ranging between −3.3‰ and

+2.4‰ PDB, with only one analysis yielding 4.0‰ PDB and (c) occurrence of a plateau around 2‰ PDB for most of the section. These features strongly resemble the global C-isotopic curve from the late Mesoproterozoic to early Neoproterozoic (Tonian), between 1,300 and 800 Ma (Kah et al., 1999; Bartley et al., 2007). Early Mesoproterozoic (>1,300 Ma) carbonates show isotopic invariance around 0‰ PDB, and late Neoproterozoic (Cryogenian-Ediacaran) successions are characterised by rapid, large-amplitude isotopic excursions and heavy δ^{13}C values between 4‰ and 12‰ PDB (Kah et al., 1999; Halverson et al., 2005; Bartley et al., 2007). A similar age around the Meso-Neoproterozoic boundary is inferred by Santos et al. (2000) for the Paranoá Group of central Brazil, which shows similar δ^{13}C values and a stromatolite assemblage also dominated by *Conophyton*.

4.4.2.2. Arroyo del Soldado Group

The thick and pure carbonate deposits of the Polanco and Cerro Victoria formations of the Arroyo del Soldado Group (ASG) provide an ideal record of the isotopic fluctuations of coeval seawater. Thus far, the database comprises 175 δ^{13}C and δ^{18}O analyses on carbonates, and a dozen sections have been studied (Boggiani, 1998; Kawashita et al., 1999a; Gaucher, 2000; Gaucher et al., 2003, 2004c,d, 2007c). Preliminary δ^{13}C analyses on organic matter from the ASG were presented by Velásquez et al. (2007, 2008).

Whereas moderate to large-amplitude δ^{13}C oscillations (up to 10‰ PDB) occur in the lower and middle ASG, low-amplitude variations (2‰ PDB) characterise the upper ASG (Gaucher et al., 2007c; Figure 4.4.2). Four positive and negative δ^{13}C excursions occur in the ASG, which are described below (Figures 4.4.2 and 4.4.3). Basinal sections of the ASG (Polanco Formation) record more negative values than proximal sections (Gaucher et al., 2004c), suggesting a strong C-isotopic gradient in the Ediacaran consistent with a stratified ocean (Calver, 2000; Frimmel, 2004; Shen et al., 2005).

4.4.2.2.1. Upper Yerbal-Lower Polanco P_1 positive excursion

This is a long-lived feature, encompassing more than 1,000 m of section (Gaucher et al., 2004c; Figures 4.4.2 and 4.4.3). A steady rise from 1‰ to 5.6‰ PDB is observed, which is concomitant to sea-level rise, shift from siliciclastic to carbonate deposits and climate warming, as also indicated by changes in clay-mineral assemblages (Gaucher, 2000; Pamoukaghlian et al., 2004). The transition to negative values up section is rather abrupt and occurs in less than 70 m (Figure 4.4.3), in which δ^{13}C values drop from +5‰ to −4.5‰ PDB.

4.4.2.2.2. Middle Polanco N_1 negative excursion

This prominent feature comprises 200 m of carbonates of unit B of the Polanco Formation (Gaucher et al., 2004c; Figure 4.4.3). The most negative values of this excursion were given as −3.3‰ PDB by Gaucher et al. (2004c) (Figure 4.4.3), but recently acquired data show even more depleted carbonates with δ^{13}C of −4.5‰ PDB. Thus, because of both the magnitude of the δ^{13}C drop and its significant stratigraphic expression, this negative excursion is the most important in the ASG. Sea-level drop, reduction of organic matter content (TOC) of carbonates, a drop in ^{87}Sr/^{86}Sr ratios (see below; Figure 4.4.3) and diminished acritarch diversity are associated to excursion N_1, suggesting that it represents a non-snowball glacial event (Gaucher et al., 2004c,d).

4.4.2.2.3. Upper Polanco P_2 positive excursion

Positive δ^{13}C values of up to 3.3‰ PDB return in units C−E of the Polanco Formation, comprising more than 300 m of section (Figures 4.4.2 and 4.4.3). Gaucher et al. (2004c) proposed that a small negative excursion may be recorded in unit D, but later work showed that δ^{13}C values never reach negative values but remain around 0‰ to 0.5‰ PDB. Therefore, this positive excursion has a central indentation (Figure 4.4.3), as also observed for feature N_1. Sea-level rise, higher TOC and higher ^{87}Sr/^{86}Sr ratios characterise carbonates of P_2 (Figure 4.4.3).

4.4.2.2.4. Terminal Polanco-Barriga Negra negative excursion N_2

A significant negative excursion marks the transition between carbonates of the Polanco Formation and carbonate breccias and conglomerates of the Barriga Negra Formation (Gaucher, 2000; Gaucher et al., 2004c). δ^{13}C values drop to −1.8‰ PDB (Figure 4.4.2), and in the shallower areas shelf exposure and erosion characterise this interval, the Barriga Negra Formation being directly derived from underlying sedimentary rocks. A significant, regional palaeokarst occurs on top of the Polanco Formation, known as the Barker Surface (Barrio et al., 1991; see Chapters 4.2 and 4.5), as a result of possibly glacioeustatic sea-level drop. In deeper settings, however, the shelf was not exposed and shales of the Cerro Espuelitas Formation directly overlie the Polanco Formation (Gaucher,

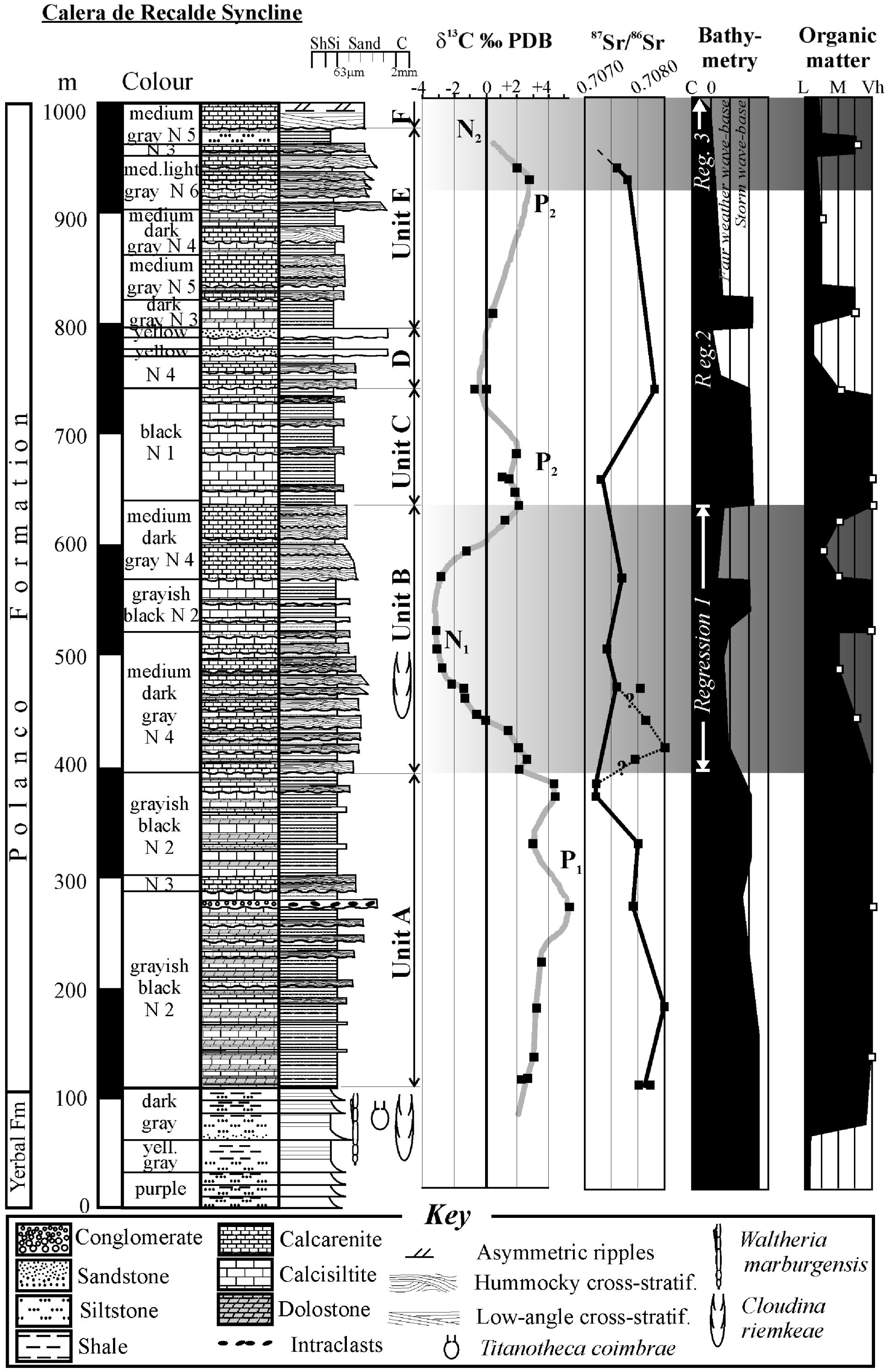

Figure 4.4.3 C and Sr chemostratigraphy and lithostratigraphy of the Polanco Formation at Calera de Recalde Syncline, modified from Gaucher et al. (2004c,d). Inferred bathymetry (C: continental; 0: swash zone) and semi-quantitative organic matter content (L: low; M: medium; Vh: very high) of carbonates is shown.

2000). There, carbonates interbedded at the base of the Cerro Espuelitas Formation also show negative $\delta^{13}C$ values, thus recording N_2 feature in a deeper setting (Gaucher et al., 2004c).

4.4.2.2.5. Cerro Espuelitas positive excursion P_3 and basal Cerro Victoria negative excursion N_3

Carbonates are very rare between the lower Cerro Espuelitas/lower Barriga Negra Formation and the lower Cerro Victoria Formation. One carbonate bed of the middle Cerro Espuelitas Formation at its stratotype yielded

δ^{13}C of 2.4‰ PDB (Figure 4.4.2). Microfossils of the Cerro Espuelitas Formation allow to place it in the Ediacaran (Gaucher, 2000), thus showing that the negative excursion at its base is older than the basal Cambrian negative excursion (Derry et al., 1994; Amthor et al., 2003; Maloof et al., 2005). The latter is likely represented in the ASG by the negative excursion at the base of the Cerro Victoria Formation, as shown by trace fossils occurring in that unit (Sprechmann et al., 2004; Gaucher et al., 2007c; Figure 4.4.2). Therefore, negative excursions N_2 and N_3 are of different age, the positive, uppermost Ediacaran δ^{13}C excursion in between (P_3) being well represented in the Nama Group (Grotzinger et al., 1995), in the Corumbá Group (Misi et al., 2007) and elsewhere.

4.4.2.2.6. Cerro Victoria positive (P_4) and negative (N_4) excursions

After ca. 130 m of carbonates yielding negative δ^{13}C values, slightly positive values around 0.3‰ PDB are recorded at the transition between units B and C of the Cerro Victoria Formation (Gaucher et al., 2007c). δ^{13}C in positive excursion P_4 reaches 0.64‰ PDB and shows fairly constant values for ca. 180 m of mainly stromatolitic carbonates (Figure 4.4.2). Up section (lower unit D), very low-amplitude, high-frequency oscillations around a mean value of 0‰ PDB are recorded in *Thalassinoides*-bearing dolostones (Sprechmann et al., 2004; Gaucher et al., 2007c). Finally, increasingly negative values of up to −1.1‰ PDB (N_4 excursion) characterise uppermost carbonates of the Cerro Victoria Formation (Figure 4.4.2). The described pattern is identical to the Lower Cambrian δ^{13}C global curve between 542 and 535 Ma (lower Nemakyt-Daldyn) presented by Maloof et al. (2005). Multiple, successive negative excursions in this interval (Maloof et al., 2005), however, prevent an unambiguous assignment of negative excursions N_3 and N_4.

4.4.2.3. Sierras Bayas Group

Gómez Peral et al. (2007) report δ^{13}C analyses for carbonates and organic matter of the Villa Mónica, Loma Negra and lower Cerro Negro formations. The Sierras Bayas Group is a shallow-water succession characterised by multiple erosional unconformities which, in practical terms, means that the δ^{13}C record is fragmentary and mostly reflects periods of high sea level.

Stromatolitic dolostones of the Villa Mónica Formation record a positive δ^{13}C excursion between −1.4‰ and +2.2‰ PDB (Gómez Peral et al., 2007), being truncated at the top by an erosional unconformity (Figures 4.2.1 and 4.4.2). Consistently positive δ^{13}C values up to 4.5‰ PDB characterise carbonates of the overlying Loma Negra Formation, which are truncated by the Barker palaeokarst (see Chapters 4.2 and 4.5; Figure 4.4.2). δ^{13}C of kerogens from the same unit vary between −28‰ and −27‰ PDB, Δ^{13}C ranging between 30.5‰ and 32‰ PDB (Gómez Peral et al., 2007). Marls of the basal Cerro Negro Formation resting atop the Barker Surface also show rising, positive $\delta^{13}C_{carbonate}$ values between 3.5‰ and 4.3‰ PDB (Figure 4.4.2) and corresponding Δ^{13}C values around 32‰ PDB. However, the fact that an important erosional surface separates the Loma Negra and Cerro Negro formations shows that the mentioned positive $\delta^{13}C_{carbonate}$ values in fact represent two different excursions (correlated here to P_1 and P_3; Figure 4.4.2) and not just one.

4.4.2.4. Piedras de Afilar Formation

Limestones occur at the top of the Piedras de Afilar Formation, and yielded δ^{13}C between 5.05‰ and 5.80‰ PDB, with little variation in 10 m of section (Pamoukaghlian et al., 2006). Rather negative δ^{18}O values between −18.1‰ and −17.5‰ are best explained by the thermal effects of nearby dolerites, which probably did not alter the carbon-isotopic composition.

4.4.3. STRONTIUM-ISOTOPE CHEMOSTRATIGRAPHY

Strontium-isotope data are available for carbonates of the ASG (Kawashita et al., 1999a; Gaucher et al., 2004d, 2007c) and Sierras Bayas Group (Kawashita et al., 1999b). In this work, we report ^{87}Sr/^{86}Sr analyses of limestones from the Mina Verdún Group.

4.4.3.1. Mina Verdún Group

Ten analyses of ^{87}Sr/^{86}Sr ratios were carried out on limestones of the El Calabozo and lower Gibraltar formations at their stratotype. Six analyses from stromatolitic, pure limestones of the El Calabozo Formation yielded values between 0.7081 and 0.7189. Four analyses of limestones from the lower Gibraltar Formation at the same section yielded very radiogenic ^{87}Sr/^{86}Sr ratios mostly between 0.7153 and 0.7174. All values are too high, and obviously reflect either post-depositional resetting and/or input of radiogenic strontium from silicates. Only the least altered samples from the El Calabozo Formation, yielding ratios of 0.7081 and 0.7084, could be regarded as near-seawater values. However, according to Shields (2007a) and Halverson et al. (2007a), seawater $^{87}Sr/^{86}Sr$ ratios ranged between 0.7050 and 0.7065 in the upper Mesoproterozoic-Tonian (1,300−800 Ma). Therefore, the currently available $^{87}Sr/^{86}Sr$ data for the Mina Verdún Group cannot be used to constrain its depositional age.

4.4.3.2. Arroyo del Soldado Group

A total of 47 $^{87}Sr/^{86}Sr$ analyses are known for the ASG, 33 being from limestones of the Polanco Formation (Gaucher et al., 2004d, 2006, this work) and 14 from dolostones of the Cerro Victoria Formation (Gaucher et al., 2007c). The behaviour of the Sr-isotopic system in these two units differ markedly from each other. Whereas dolostones of the Cerro Victoria Formation yield too radiogenic values (>0.7106: Gaucher et al., 2007c), limestones of the Polanco Formation yield mostly near-primary $^{87}Sr/^{86}Sr$ ratios (Figure 4.4.1C). This is readily explained by the lower Sr concentrations that dolomite allows in its lattice, which renders it more susceptible to post-depositional alteration (Gaucher et al., 2007c). On the other hand, limestones of the Polanco Formation are characterised by high Sr concentrations of up to 2,250 ppm (mean = 1,025 ppm, standard deviation = 470 ppm and $N = 88$), and Rb concentrations typically less than 10 ppm, denoting a negligible proportion of radiogenic Sr from silicates. $^{87}Sr/^{86}Sr$ and $\delta^{13}C$ crossplots versus common alteration proxies (Figure 4.4.1C) show the unaltered nature of these isotopic signatures. Considering this, only the results from the Polanco Formation will be discussed, which are the ones useful for strontium chemostratigraphy.

All samples from the Polanco Formation show $^{87}Sr/^{86}Sr$ values within 0.7073 and 0.7086, the only exceptions being samples near the contact of the intrusive Minas Granite, with values up to 0.7156. Even there, one sample retained a near-primary value of 0.7077 (Gaucher et al., 2006). The variation of $^{87}Sr/^{86}Sr$ ratios in limestones of the Polanco Formation is not random, but shows secular variations of an amplitude of 0.0010 (Gaucher et al., 2004d; Figure 4.4.3). The lowermost Polanco Formation is characterised by declining $^{87}Sr/^{86}Sr$ ratios between 0.7085 and 0.7080 (Figure 4.4.3), dropping to a nadir of 0.7073 at peak $\delta^{13}C$ values of the uppermost unit A (Gaucher et al., 2004d). Throughout units B and C, $^{87}Sr/^{86}Sr$ values remain low (0.7073−0.7077), only increasing in unit D to a maximum of 0.7083. One possible exception could be the apparent surge of $^{87}Sr/^{86}Sr$ values from 0.7073 to 0.7086 at the base of unit B at Calera de Recalde Syncline (Figure 4.4.3), but this needs confirmation from other sections. A final drop to 0.7076 is observed at the top of the Polanco Formation (unit E; Figure 4.4.3).

The consistency of the observed pattern, along with the geochemical indications of a primary isotopic signature (Figure 4.4.1), suggest that the observed $^{87}Sr/^{86}Sr$ oscillations reflect secular variations of seawater composition. This has profound implications for both Neoproterozoic Sr-isotope chemostratigraphy and palaeoclimate models (see below).

4.4.3.3. Sierras Bayas Group

Gómez Peral et al. (2007) show, on the basis of different geochemical and isotopic proxies, that limestones of the Loma Negra Formation retain primary C- and Sr-isotopic signatures. Kawashita et al. (1999b) reports $^{87}Sr/^{86}Sr$ values between 0.7069 and 0.7087 for the Loma Negra Formation, with the least altered values between 0.7069 and 0.7075. As noted by Gaucher et al. (2005b), the $^{87}Sr/^{86}Sr$ ratios of the Loma Negra Formation are almost identical to the ones reported for the Polanco Formation of the ASG. Both units were previously correlated on the basis of litho- and biostratigraphy (Gaucher et al., 2005b; see Chapters 4.2 and 4.3; Figure 4.4.2).

No $^{87}Sr/^{86}Sr$ analyses are available for dolostones of the Villa Mónica Formation, which usually contain less than 45 ppm Sr (Gómez Peral et al., 2007).

4.4.4. SYNTHESIS

The age of the Mina Verdún Group is still poorly constrained, but the $\delta^{13}C$ plateau around 2‰ PDB observed in the El Calabozo and Gibraltar formations is typical of late Mesoproterozoic to Tonian carbonates (1,300−800 Ma: Bartley et al., 2007). U-Pb dating of interbedded acid volcanics described by Gaucher et al. (2007a) will help improve the still poorly constrained pre-Cryogenian global $\delta^{13}C$ curve.

Poiré and Gaucher (2007) pointed out that dolostones of the Villa Mónica Formation show a $\delta^{13}C$ curve similar to carbonates of the Mina Verdún Group, both units being pre-Ediacaran in age and containing *Conophyton* stromatolites (Figure 4.4.2). However, similar, leiosphere-dominated acritarch assemblages in the Villa Mónica

Formation and overlying units of the Sierras Bayas Group militate against this interpretation (Gaucher et al., 2005b; see Chapter 4.3). Thus, the issue of the correlation of the Villa Mónica Formation remains controversial.

The ASG contains without doubt the most detailed late Ediacaran chemostratigraphic record in whole southwestern Gondwana between ca. 570 and 550 Ma. Positive excursion P_1 is recorded in the Yerbal and Polanco formations in the ASG, and in the Loma Negra Formation in the Sierras Bayas Group (Figure 4.4.2). In both units, *Cloudina riemkeae* and low-diversity acritarch assemblages occur (Gaucher et al., 2005b; see Chapter 4.3). The impressive negative excursion N_1 that follows may be correlated to the Shuram–Wonoka anomaly (Halverson et al., 2005), which ended at 551 ± 1 Ma and reached its nadir before 555 ± 6 Ma in south China (Condon et al., 2005; Zhang et al., 2005). $\delta^{13}C$ values in the Shuram–Wonoka anomaly are as low as $-10‰$ PDB or less (Halverson et al., 2005), but in the Polanco Formation $\delta^{13}C$ does not drop beyond $-4.5‰$ PDB. This, however, can be due to ocean stratification or other local effects (Calver, 2000; Frimmel, 2004; Shen et al., 2005). The sedimentary record of the Polanco Formation favors a Phanerozoic-type glaciation as the cause of the negative anomaly (Gaucher et al., 2004d; Gaucher, 2007; see Chapter 4.5), as also observed for other Neoproterozoic negative anomalies. Most interestingly, $^{87}Sr/^{86}Sr$ reaches minimum values before the onset of N_1, and stays low (0.7075) well into the following positive excursion (P_2; Figures 4.4.2 and 4.4.3). The decrease of $^{87}Sr/^{86}Sr$ values during a glacial event could be due to: (a) ice cover of continents, diminishing the input of ^{87}Sr to the oceans (Jacobsen and Kaufman, 1999); (b) pre-glacial, enhanced hydrothermal activity in mid-ocean ridges leading to plankton blooms, CO_2 sequestration and glaciation (Gaucher, 2007); and (c) a combination of both. Considering that $^{87}Sr/^{86}Sr$ ratios begin to fall well before the onset of negative anomaly N_1 (Figure 4.4.3), enhanced hydrothermal activity linked to rifting events seems a more attractive hypothesis.

The same sequence of events is observed during the terminal Polanco-Barriga Negra negative excursion N_2, which predates platform exposure in both the Arroyo del Soldado and the Sierras Bayas Group (Figure 4.4.2), leading to the regionally important Barker palaeokarst (Barrio et al., 1991; Poiré and Gaucher, 2007). A tentative correlation of this surface with the Vingerbreek Member in the Nama Group, constrained between 549 ± 1 and 545 ± 1 Ma (Grotzinger et al., 1995; Saylor et al., 1998), showing platform reworking, a negative $\delta^{13}C$ excursion to $-2‰$ PDB and evidences of a glacial event (Germs, 1995) is considered plausible (see Chapter 4.5).

Carbonates of the Cerro Victoria Formation are confidently assigned to the Lower Cambrian on the basis of trace fossils and carbon isotopes (Gaucher et al., 2007c, and references therein). The $\delta^{13}C$ curve reported for this unit is identical to the interval between 542 and 535 Ma (lower Nemakyt-Daldyn) in the high-resolution global $\delta^{13}C$ curve presented by Maloof et al. (2005). This is firm evidence that coeval Cambrian carbonates in the Cuyania-Precordillera Terrane in Argentina have potential counterparts in core areas of Gondwana (Finney, 2007), apart from Laurentia. The tectonic evolution of the area may explain the absence of carbonates – or other sedimentary rocks – younger than 535 Ma in the RPC (see Chapter 4.6).

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