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SUPPLEMENTARY GEOLOGICAL BACKGROUND

Callison Lake Formation, Yukon, Canada

The Callison Lake Formation of the Mount Harper Group (Windermere Supergroup) is widely exposed in the Coal Creek and Hart River inliers of the Ogilvie Mountains, Yukon, Canada (Fig. 1, main text). These strata have been subdivided into four informal members (Heterolithic, Talc, Ramp, and Transitional), which reflect different phases of mixed siliciclastic and carbonate sedimentation in a subtropical, restricted to open marine extensional basin formed during the break-up of Rodinia (Strauss et al., 2015). The Callison Lake Formation is bracketed in age between ca. 811 and 719 Ma by U-Pb chemical abrasion-isotope dilution-thermal ionization mass spectrometry (CA-ID-TIMS) dates on zircon from volcanic horizons in bounding strata (Fifteenmile Group and Mount Harper volcanic rocks, respectively) (Macdonald et al., 2010, 2018). Callison Lake strata are also temporally contained by two Re-Os depositional ages on organic-rich mudstones: 1) a 752.7 ± 5.5 Ma date from the Heterolithic-Talc member contact (Rooney et al., 2015), and 2) a 739.9 ± 6.1 Ma date from the middle portion of the Transitional member (Fig. 1, main text; Strauss et al., 2014), making it one of the well-calibrated Tonian successions in the world.

Mg-silicates in the Callison Lake Formation occur as meter- to decameter-thick accumulations in the ~45–109 m-thick Talc member (Tosca et al., 2011; Strauss et al., 2015). These strata are mostly characterized by talc-rich black mudstone and diverse microbial dolostone facies deposited within a subtidal to supratidal depositional environment (Strauss et al., 2015). The Mg-silicates form stratiform cm- to m-thick intervals of talc-rich mudstone, drapes on stromatolitic laminae, and a wide variety of primary coated grains, such as ooids, oncolites, pisoids, and cortoids (Fig. 1, main text). Talc-rich intervals are occasionally capped by erosional surfaces where talc-rich mudstone is ripped-up to form cross-bedded intraclast dolowackestone or dolopackstone. The presence of sulfate evaporite pseudomorphs, tepees, mudcracks, and the intimate association of microbial dolostone and organic-rich fine-grained strata all indicate that Talc member strata were deposited in a periodically flooded tidal flat or sabkha (Strauss et al., 2015). A marine setting for these strata is also indicated by m-scale shoaling-upwards parasequences, as well as the presence of globally-recognized vase-shaped microfossils (Strauss et al., 2014; Cohen et al., 2017a) and unradiogenic Os_i isotopic data in Talc member mudstone (Strauss et al., 2014; Rooney et al., 2015).

Mg-silicates are absent in other siliciclastic and carbonate strata of the Callison Lake Formation, except in the form of rare drapes along stromatolitic bioherms in the Heterolithic and Transitional members (Strauss et al., 2015). Carbonate strata throughout the Callison Lake Formation are more commonly characterized by eponymous Neoproterozoic microbial fabrics, such as trap-and-bind stromatolites, microbialites, and oncolites, as well as thick accumulations of laminated to massive dolomudstone, dolowackestone, and dolograinstone dominated by ooids, intraclasts, and pisoids (Strauss et al., 2015). Despite evidence for evaporative carbonate sedimentation in the Callison Lake Formation, carbonate strata do not host evidence for precipitated fabrics, such as aragonite fans or microdigitate stromatolites, nor are there any notable accumulations of CaCO₃ microspar cements or molar-tooth structures.

46 ***Svanbergfjellet Formation, Nordhaustlandet and Ny Friesland, Svalbard, Norway***

47 Neoproterozoic rocks exposed in northeastern Svalbard are part of the upper Hecla Hoek
48 Series of the Northeastern basement province (Harland and Wilson, 1956; Wilson, 1958, 1961;
49 Dallmann, 2015). The Hecla Hoek Series outcrop in nunataks and coastal sections in northeastern
50 Spitsbergen and northwestern Nordaustlandet and is subdivided into the Veteranen,
51 Akademikerbreen, and Polarisbreen groups (Fig. 1, main text). Whereas a different nomenclature
52 has historically been used for strata exposed on the island of Nordaustlandet (Flood et al., 1969),
53 correlation of these strata across Hinlopenstretet is unambiguous, and it has become the convention
54 to use the better-known stratigraphic nomenclature from Spitsbergen when describing
55 Neoproterozoic sedimentary rocks in northeastern Svalbard (Halverson et al., 2018a). This is
56 important because Tosca et al. (2011) previously described Mg-silicates from the Hunnberg
57 Formation (Flood et al., 1969), which is correlative with the Svanbergfjellet Formation (Fig. 1,
58 main text; Harland and Wilson, 1956; Wilson, 1958, 1961; Knoll and Swett, 1990).

59 The ~2 km thick Akademikerbreen Group (Tonian) is almost exclusively composed of
60 carbonate, and has been subdivided into the Grusdievbreen, Svanbergfjellet, Draken, and
61 Backlundtoppen formations. The upper Grusdievbreen and lower Svanbergfjellet formations
62 record the entirety of the ca. 810–800 Ma Bitter Springs carbon isotope excursion (Halverson et
63 al., 2007, 2018a; Macdonald et al., 2010; Swanson-Hysell et al., 2015; Cohen et al., 2017b). The
64 overlying Polarisbreen Group (specifically, the Russøya Member) preserves a negative $\delta^{13}\text{C}$
65 anomaly that was previously correlated with the so-called ca. 735 Ma Islay anomaly of Scotland
66 (Hoffman et al., 2012; Rooney et al., 2014; Strauss et al., 2014; MacLennan et al., 2018). The basal
67 Ediacaran Dracöisen cap dolostone occurs across the entire outcrop belt and serves as a useful
68 chronostratigraphic marker (Harland et al., 1993; Halverson et al., 2004) that can be confidently
69 assigned an age of ca. 635 Ma (Hoffmann et al., 2004; Condon, 2005; Calver et al., 2013).

70 The carbonate-dominated ca. 805–788 Ma Svanbergfjellet Formation (Halverson et al.,
71 2018b) is subdivided into four members, which in stratigraphically-ascending order consist of the
72 ~150 m-thick Lower Dolomite, ~150 m-thick Lower Limestone, ~120–150 m-thick Algal
73 Dolomite, and ~70–150 m-thick Upper Limestone (Knoll and Swett, 1990). Dolomite-dominated
74 strata of the Lower and Algal Dolomite members are characterized by interbedded stromatolitic
75 boundstone, dolomudstone, intraclast packstone and wackestone, and dolograstone with
76 subordinate intervals of black to maroon mudstone. These strata record subtidal to peritidal
77 sedimentation in a tidal flat to lagoonal depositional environment, as indicated by a combination
78 of subaerial exposure indicators, such as mudcracks and tepees, as well as subtidal indicators
79 including stromatolitic buildups, cross-bedded oolitic grainstones, and finely laminated carbonate
80 and siliciclastic mudstones (Knoll and Swett, 1990; Maloof et al., 2006; Halverson et al., 2007).
81 In contrast, limestone-dominated members of the Svanbergfjellet Formation are composed of dark
82 grey to black, thin-bedded and planar- to wavy-laminated lime mudstone with abundant molar-
83 tooth structures, which mostly reflect subtidal sedimentation in a shallow shelf to lagoonal
84 depositional environment (Knoll and Swett, 1990).

85 Tosca et al. (2011) described nodular Mg-silicate occurrences associated with void-filling
86 CaCO_3 microspar cements in stromatolitic and molar-tooth structures from both the Algal
87 Dolomite and Upper Limestone members, which were previously misinterpreted as phosphate
88 nodules (Knoll, 1984; Knoll and Swett, 1990). Our more recent observations of strata exposed at
89 Roaldtoppen in Murchisonfjorden, Nordhaustlandet (Fig. DR1), suggest that Mg-silicates are also
90 preserved in the Lower Dolomite Member, both in the form of cm- to m-thick talc-rich black
91 mudstone deposits, as well as nodular and stratiform aggregates associated with CaCO_3 microspar

in lime mudstone, stromatolitic horizons, and molar-tooth structures (Fig. 2, main text; Fig. DR1). These Mg-silicate-rich strata are commonly reworked by current-generated structures, such as trough cross-stratification and hummocky cross-stratification, suggesting they formed at or near the sediment-water interface. Finally, Mg-silicates in the lower Svanbergfjellet Formation are also preserved as a wide variety of coated grain lithofacies, similar to what is described above from the Callison Lake Formation.

On the Relationship Between Mg-Silicate and CaCO₃ Microspar Authigenesis

Tosca et al. (2011) provided experimental evidence for the mechanisms of Mg-silicate nucleation and crystallization from seawater-derived fluids, in which a primary amorphous Mg-silicate phase undergoes crystallization and maturation (accompanied by loss of structural H₂O) to generate sedimentary talc. Because talc (in addition to other Mg-silicates) is hydrophobic, its presence in aqueous systems would be predicted to generate immiscible textures; that is, physical separation of the liquid phase from the adjacent hydrophilic minerals. Consistent with this observation, nodules documented from the Svanbergfjellet Formation exhibit “oil-in-water” textures, where Mg-silicate nodules are surrounded by CaCO₃ microspar, as evidenced by the aggregation and physical separation of Mg-silicate crystallites from the surrounding microcrystalline CaCO₃ matrix (Fig. 2D-F, main text). This provides direct support that Mg-silicate nodules were initially precipitated as fine-grained crystallites in intimate contact with microcrystalline CaCO₃, and that they rapidly aggregated together in a CaCO₃ matrix that was fluid (and thus not fully lithified). In addition, this indicates that the Mg-silicates and CaCO₃ microspar precipitated contemporaneously, and that the two phases physically separated due to surface charge disparities. This is inconsistent with alternative models for nodule growth where the diffusion of components within the sediment either displaces or replaces the sediment matrix (in this case CaCO₃ microspar). On the contrary, the microspar matrix surrounding the Mg-silicates exhibits no evidence for displacement or replacement and is composed instead of euhedral, interlocking crystalline masses.

Although Tosca et al. (2011) initially interpreted the Mg-silicate precipitates as a wholly diagenetic phenomenon, their explanation is at odds with mass balance constraints, as well as geochemical and sedimentological data. First, as stated in the manuscript, the presence of pore-filling CaCO₃ microspar requires the delivery of significant amounts of seawater through the uppermost portion of the sediment. If we assume that seawater [Ca] was equivalent to ~10 mmol/kg (i.e., Spear et al., 2014), and assume that only 50% of the CaCO₃ microspar volume was originally precipitated (and the remaining 50% precipitated as a later overgrowth), a microspar-rich ribbon 7 x 5 x 1 cm in size requires more than 47 liters of seawater to form. This strongly supports the inference that synsedimentary CaCO₃ microspar cement, like any other syndepositional carbonate cement, reflects crystallization from seawater rather than diagenetic fluids, a conclusion reached by many previous authors, including Shields (2002), Bishop and Sumner (2006), and James et al. (1998). Bishop and Sumner (2006) proposed that large amounts of seawater could be injected into permeable and porous sediments through hydrostatic responses to wave oscillation (i.e., storms or normal wave activity above fair-weather wave base), which is consistent with the paleoenvironmental interpretation for the Svanbergfjellet Formation.

Previous studies of CaCO₃ microspar have also documented that microspar crystallization took place (1) before appreciable compaction of the sediment, and (2) while the sediment itself was susceptible to physical re-working. For example, the Svanbergfjellet Formation contains CaCO₃ microspar clasts and CaCO₃ microspar grainstone lags, which speaks to the erodibility of

the sediment shortly after the microspar precipitated. This phenomenon has also been observed in many CaCO_3 microspar-bearing successions (e.g., James et al., 1998).

A diagenetic origin for microspar CaCO_3 is inconsistent with geochemical observations on Tonian carbonate rocks in general (and the Svanbergfjellet Formation in particular). Although Tosca et al. (2011) noted that microbial sulfate reduction and microbial iron reduction are effective alkalinity pumps that could drive CaCO_3 supersaturation, the Svanbergfjellet carbonates, like many other examples (Grotzinger and James, 2000), are devoid of the metabolites produced by both of these processes. In other words, sedimentary Fe, in any form, as well as sulfur, are both conspicuously absent in these sediments, as is organic matter (e.g., Sperling et al., 2015; Kunzmann et al., 2017; Sperling and Stockey, 2018). Although oxic respiration of organic matter during diagenesis may influence the chemistry of pore waters and result in negligible organic matter preservation, this process consumes alkalinity rather than producing it, resulting in an overall pH decrease. These geochemical observations are also consistent with $\delta^{13}\text{C}$ analyses of microspar (e.g., Tracy and Lyons, 1998; Shields, 2002; Hodgskiss et al., 2018), which show no anomalous negative $\delta^{13}\text{C}$ fractionation, and are consistent with the $\delta^{13}\text{C}$ of contemporaneous carbonate sediments worldwide. Critically, the latter study by Hodgskiss et al. (2018) claimed that Fe reduction was a key process in creating enough alkalinity to generate molar tooth structures; however, the extreme sensitivity of the Fe isotope system to host phases was not explored in that study, which makes the isotopic results ambiguous. Together, mass balance constraints, in combination with geochemical and sedimentological observations, support the interpretation that pore-filling microspar cement reflects fluids dominated by contemporaneous seawater.

CARBONATE SYSTEM REACTION PATH MODELLING

In order to quantitatively examine the effects of variable alkalinity and $[\text{Ca}^{2+}]$ ratios in Neoproterozoic seawater on pH and mineralogy, we executed reaction path models of evaporation that take into account changing CaCO_3 solubility, CO_2 solubility, and aqueous carbonate speciation with increasing ionic strength. All evaporation models were performed with the Geochemist's Workbench v11.0 (Bethke and Yeakel, 2015) software using the Pitzer ion interaction model for calculating ion activity coefficients and mineral solubility upon evaporation (Pitzer, 1973; Harvie et al., 1984). Pitzer ion interaction parameters for the dissolved carbonate system, as well as apparent dissociation constants for H_2CO_3^0 and HCO_3^- , were taken from He & Morse (1993); however, in order to more realistically account for CaCO_3 precipitation from seawater-derived fluids, the apparent solubility of calcite in seawater was taken from Mucci (1983). Because the solubility products of aragonite and calcite are similar to one another, the choice of CaCO_3 polymorph has no discernible effect on the subsequent evolution of evaporating fluids. We chose calcite as the polymorph for modelling calculations here, acknowledging that either aragonite or calcite (or both) may have been dominant CaCO_3 phases precipitating from Neoproterozoic seawater or its derivatives. Evaporation models were run by specifying equilibrium between atmospheric CO_2 and dissolved carbonate species in solution (Lazar et al., 1983). Back reaction between minerals and the evolving fluid was permitted. All simulations begin with concentrations for Na^+ , K^+ , Mg^{2+} , Cl^- , and SO_4^{2-} constrained by Spear et al. (2014). Atmospheric CO_2 concentration (and therefore total alkalinity), pH, and Ca^{2+} concentrations were adjusted to examine situations where $2*[\text{Ca}^{2+}]$ was less than or greater than the total alkalinity, keeping estimates of these parameters consistent with constraints presented the main text.

The results of the modelling simulations show that the pH threshold of 8.5, required to nucleate Mg-silicate directly from Neoproterozoic seawater (Tosca et al., 2011), is only crossed

when $[ALK] > 2*[Ca^{2+}]$. Under these conditions (Fig. DR2), pH increases to a maximum value of 8.62, and remains above pH 8.5 during and after $CaCO_3$ precipitation and through the precipitation of halite (NaCl). When $[Ca^{2+}]$ is quantitatively consumed by $CaCO_3$ precipitation under these conditions, gypsum ($CaSO_4 \cdot 2H_2O$) is not predicted as a significant direct precipitate from water (Fig. DR3); however, modelling results show that late-stage evaporation leads to the precipitation of anhydrite and, in some cases, the back-reaction of $CaCO_3$ and the subsequent formation of gypsum or anhydrite. This implies that gypsum and/or anhydrite ($CaSO_4$) are not exclusively prohibited from forming in systems where $[ALK] > 2*[Ca^{2+}]$, which is consistent with sedimentological observations from the Callison Lake Formation (Strauss et al., 2015). $CaCO_3$ back-reaction is due to the effect of calcite solubility changes with increasing evaporation and the subsequent release of Ca^{2+} which can promote gypsum precipitation. These late-stage effects are also consistent with studies of Lazar et al. (1983) and McCaffrey et al. (1987), but it is worth noting that other geochemical factors may also influence the presence or absence of gypsum/anhydrite formation, as well as the concentration factor associated with its formation. For example, increases in $[SO_4^{2-}]$ beyond the 3 mmol/kg used here (Spear et al., 2014) result in earlier and more abundant gypsum/anhydrite precipitation, independent of the relationship between $[ALK]$ and $[Ca^{2+}]$.

Reaction path modelling results for Tonian seawater confirm that when $[ALK] < 2*[Ca]$ (Fig. DR2), pH decreases upon calcite precipitation, similar to modern seawater (Krumgalz et al., 1980; Lazar et al., 1983; McCaffrey et al., 1987). Under these conditions, the bulk mineralogy includes calcite, gypsum, halite, and late stage chloride salts. The modelling results show that when Ca^{2+} is not qualitatively depleted by $CaCO_3$ precipitation, gypsum and halite precipitation occur almost simultaneously at concentration factors of approximately 10x, which is consistent with constraints from Spear et al. (2014). Building upon the results of these constraints, we performed carbonate chemistry calculations using custom scripts written in MATLAB for both the Callison Lake Formation (Fig. DR4) and Svanbergfjellet Formation (Fig. DR5), which are described further in the main text.

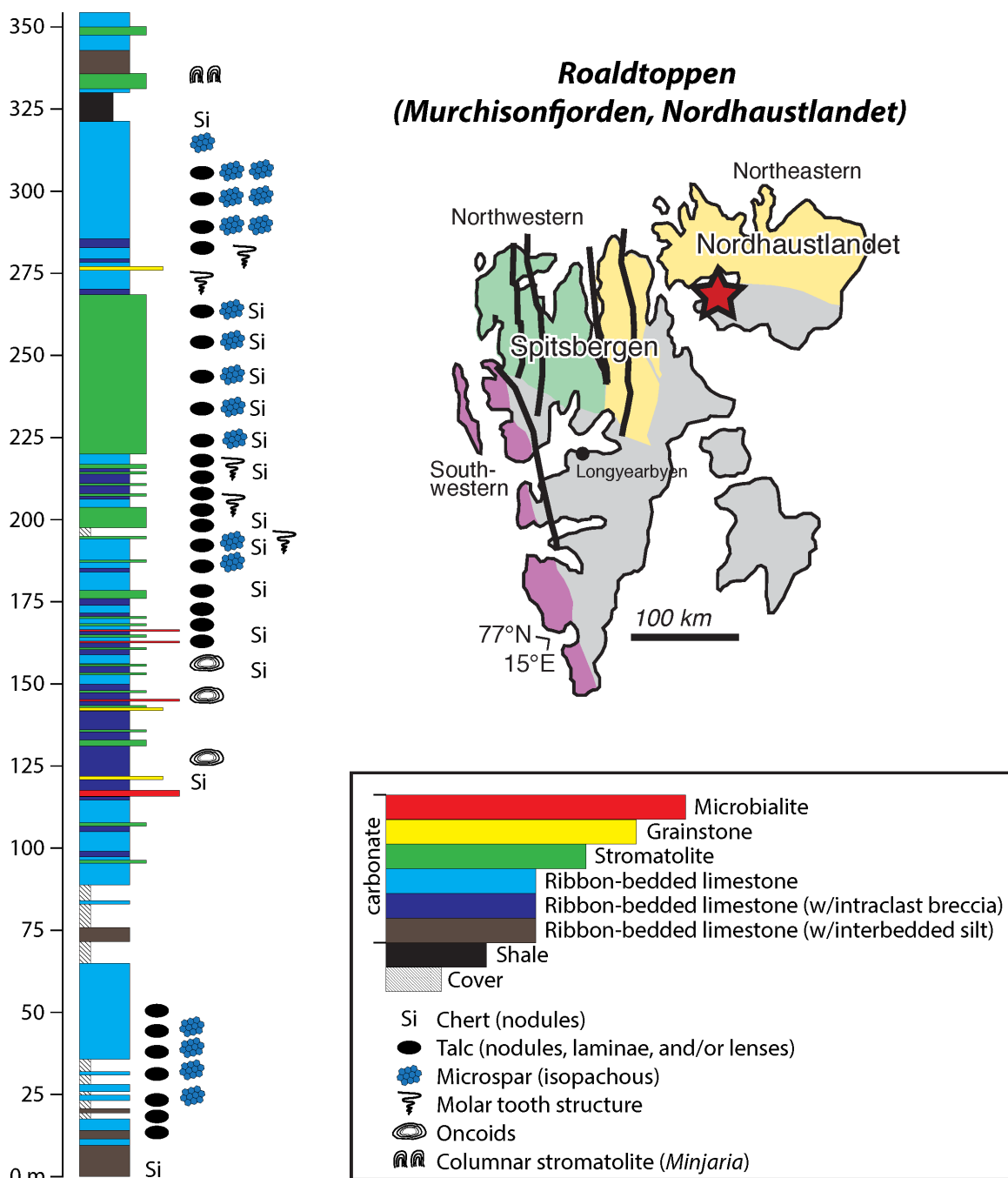


Figure DR1. Measured section (left) of the lower Svanbergfjellet Formation (Lower Dolomite Member) from Roaldtoppen, Murchisonfjorden, Nordhaustlandet, Svalbard. Inset map (upper right) shows the location of the measured section (red star) and legend (lower right) describes the main observed lithofacies within the measured section. Note the widespread occurrence of Mg-silicates (talc) and CaCO_3 microspar throughout the measured section. Petrographic thin sections presented in Fig. 2 of the main text are from ~175-180 m in the measured section.

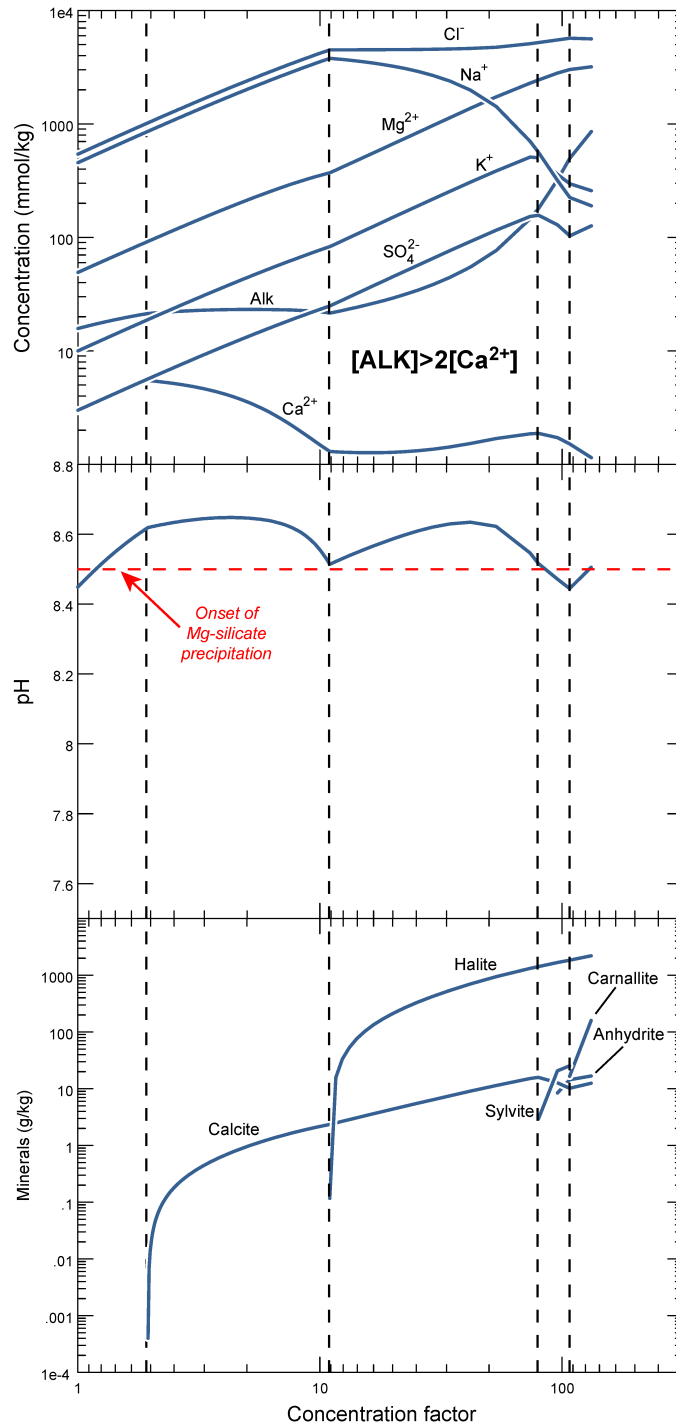


Figure DR2. Reaction path model outputs for the evaporation of Neoproterozoic seawater (based on the composition reported in Spear et al., 2014) where $[ALK] > 2[Ca^{2+}]$. Under these conditions, Ca^{2+} is rapidly consumed during chemically induced $CaCO_3$ precipitation and residual alkalinity increases with evaporation (**top**), resulting in a sustained increase in pH (**middle**). The onset of Mg-silicate precipitation, at pH 8.5, is reached close to the onset of $CaCO_3$ precipitation in this example, but well before halite precipitation (**bottom**). pH continues to increase to a maximum of 8.62.

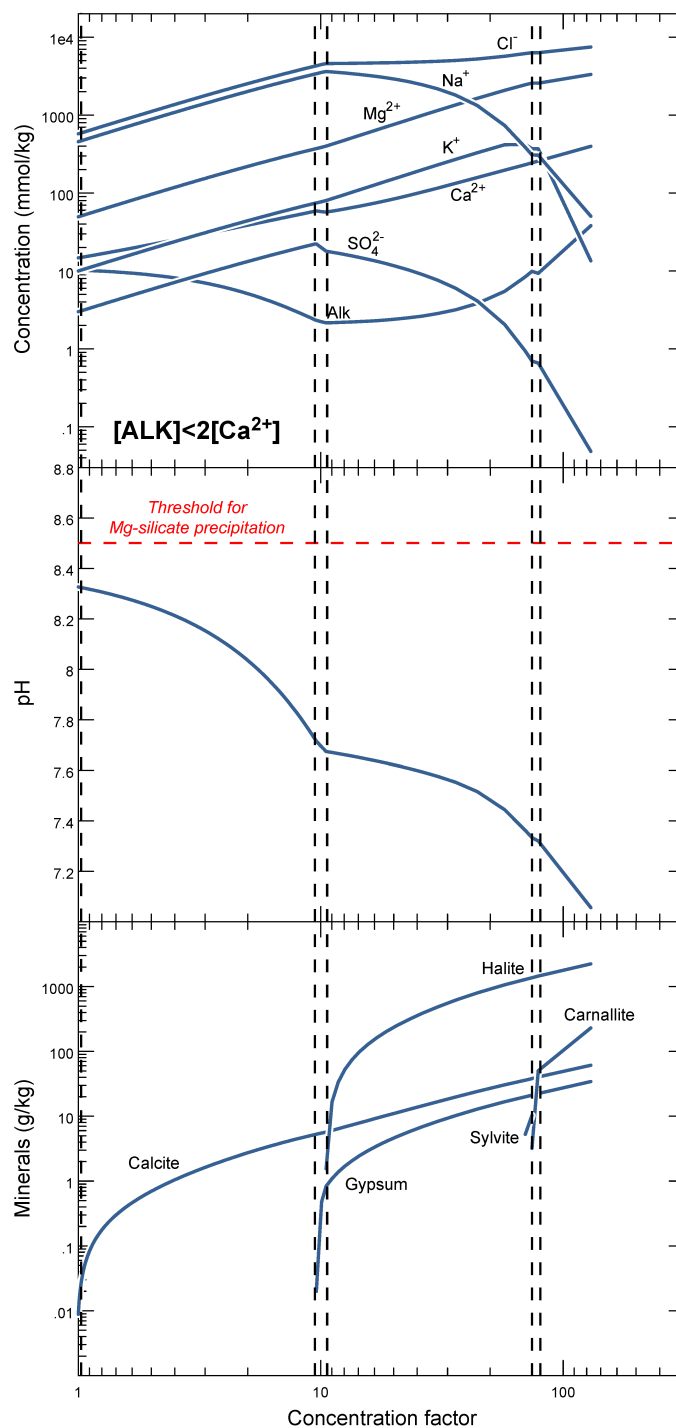


Figure DR3. Reaction path model outputs for evaporation of Neoproterozoic seawater (based on the composition reported in Spear et al., 2014) where $[ALK] < 2[Ca^{2+}]$. Under these conditions, $[ALK]$ is largely consumed through chemically induced $CaCO_3$ precipitation (**top**), which results in a decrease in pH (**middle**). Gypsum and halite both precipitate at concentration factors of ~ 10 (**bottom**), in close agreement with calculations presented by Spear et al. (2014).

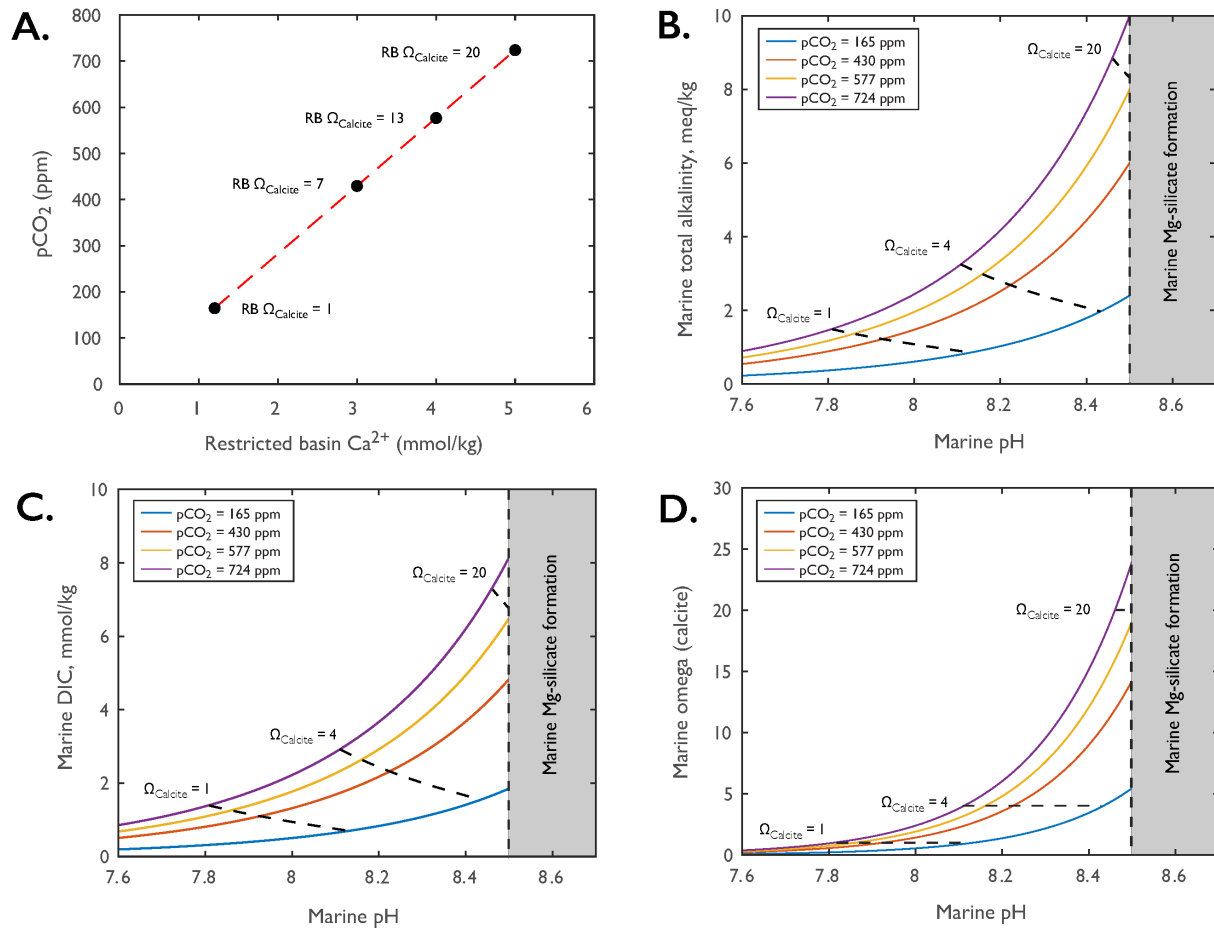


Figure DR4. Tonian carbonate equilibrium constraints derived from the ca. 755–735 Ma Callison Lake depositional system. **A)** Plot of $p\text{CO}_2$ vs. $[\text{Ca}^{2+}]$ for the Callison Lake restricted basin (RB) at different Ω_{Calcite} values. **B)** Plot of total marine alkalinity vs. pH for open marine systems at the time of Callison Lake deposition at different Ω_{Calcite} and $p\text{CO}_2$ values. **C)** Plot of total marine DIC vs. pH for open marine systems at the time of Callison Lake deposition at different Ω_{Calcite} and $p\text{CO}_2$ values. **D)** Plot of marine Ω_{Calcite} vs. pH for open marine systems at the time of Callison Lake deposition at different $p\text{CO}_2$ values. In **A–D**, the grey area defines a chemical space where marine pH would be above the critical supersaturation level for Mg-silicate authigenesis.

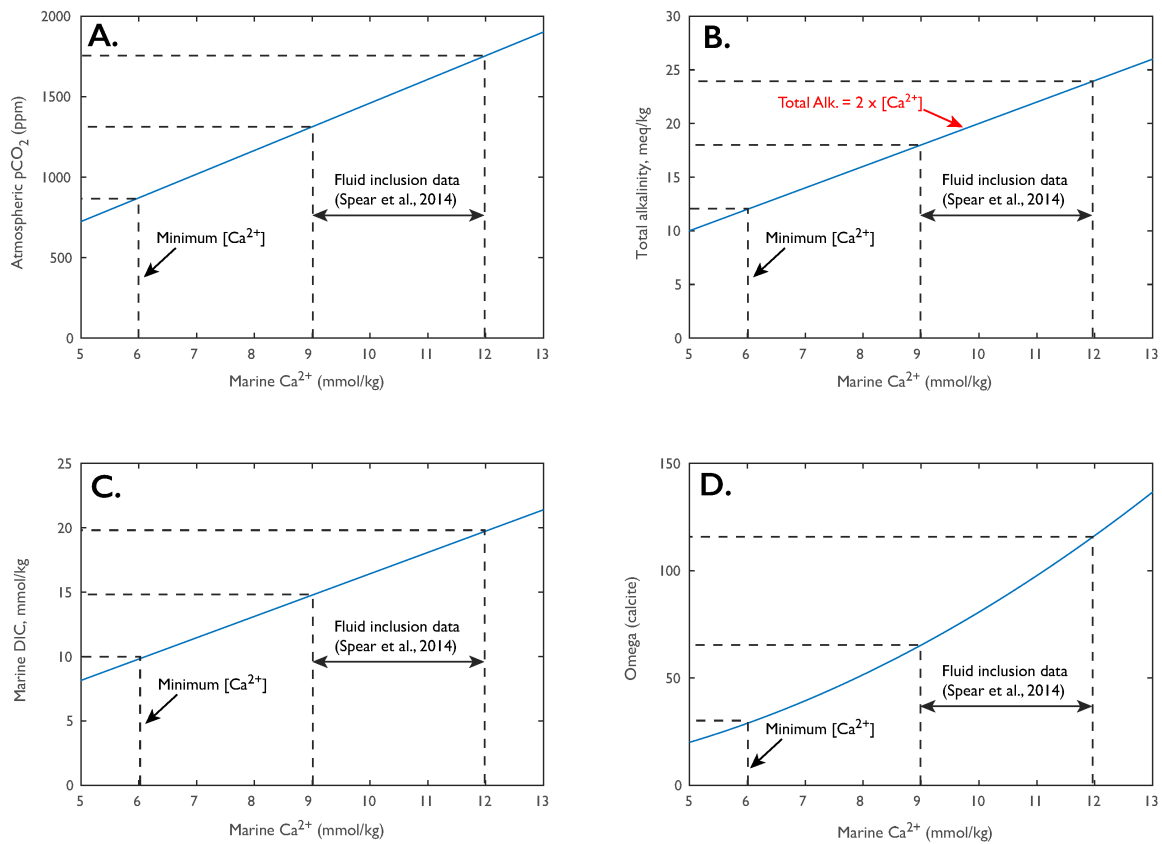


Figure DR5. Tonian carbonate equilibrium constraints derived from the ca. 805–788 Ma Svanbergfjellet Formation. **A)** Plot of atmospheric $p\text{CO}_2$ vs. $[\text{Ca}^{2+}]$ for the Svanbergfjellet Formation based on calculated Tonian $[\text{Ca}^{2+}] = 6\text{--}12$ mmol/kg from Spear et al. (2014) and minimum from evaporation modeling. **B)** Plot of total marine alkalinity vs. $[\text{Ca}^{2+}]$ for the Svanbergfjellet Formation. **C)** Plot of marine DIC vs. $[\text{Ca}^{2+}]$ for the Svanbergfjellet Formation. **D)** Plot of Ω_{Calcite} vs. $[\text{Ca}^{2+}]$ for the Svanbergfjellet Formation.

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