RESEARCH ARTICLE

Dynamics of marine inorganic carbon and silica: A field study of the mechanisms controlling seawater major element concentrations

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Abstract

A highly resolved time series of dissolved major element (calcium, strontium, magnesium, and lithium) concentrations in the north Gulf of Aqaba, Red Sea, reveals variability in major cation concentrations beyond analytic uncertainties. This variability is composed of an interannual component that is most important for calcium, and a short-term daily-timescale component that is most important for lithium. As evident from covariation in calcium, potential alkalinity, and Sr/Ca, the calcium carbonate cycle of the Gulf of Aqaba is dominated by coral calcification, and there was an increase in calcification rates between 2017 and 2018. Variability in lithium concentrations, and larger changes in magnesium concentrations than expected from magnesium distribution coefficients in carbonate minerals, suggest an active cycle of aluminosilicate mineral dissolution, and precipitation of secondary silicate minerals.

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Sea salt is primarily composed of 11 ions at nearly constant ratios; this is known as the Law of Constant Proportions (Broecker and Peng 1982; Culkin and Cox 1966; Dittmar 1884). These ions, commonly referred to as "major elements" are found in seawater at concentrations $> 1 \, \mu$mol kg^{-1}, and together make up $> 99\%$ of the soluble ionic constituents of seawater (Kremling 1999). With the exception of the major nutrients (carbon, silicic acid, nitrate, and phosphate), the major constituents of sea salt have long residence times in the ocean, ranging from 500,000 to 100,000,000 yr (Broecker and Peng 1982; Lecuyer 2016). Comparing the long residence times of major dissolved elements in seawater with the ~ 2000 yr it takes to fully mix and ventilate the ocean (Matsumoto 2007), one might expect that there should be minimal variability in major element ratios, and that major element concentrations will vary only due to water evaporation and precipitation. Even though constant ratios

are the general case for most major elements, it is established that the salinity-normalized concentrations of strontium (Sr) and lithium (Li) vary by up to 3% in the open ocean (de Villiers 1999; Steiner, Landing, et al. 2022) and salinity-normalized calcium (Ca) concentrations vary by 1.5–2% in the open ocean (Krumgalz 1982; Steiner et al. 2021). There is also evidence for higher variability in Sr/Ca and Mg/Ca of the modern ocean (Lebrato et al. 2020). The ability to observe fine-scale changes in the chemical composition of seawater improved over time thanks to increasing analytical capabilities, however, scarcity of reliable measurements of major element concentrations in the modern ocean make it difficult to assess to what degree the chemical composition of seawater can be altered by boundary processes.

Reactions along ocean boundaries such as mixing of river and seawater, interactions of seawater with aerosols and reactions occurring at and below the sediment–water interface dominate the supply and removal of many elements in the ocean (e.g., Anderson 2020; Hamilton et al. 2022; Homoky et al. 2016). In the case of major elements, mixing of seawater with water from other sources is expected to be conservative, that is, the mere act of water mixing is not expected to induce chemical reactions removing major elements. Rivers and groundwater are prominent sources of dissolved ions to the ocean, typically featuring high cation to sodium (Na) ratios (e.g., Beckwith, Byrne, and Hallock 2019; Mayfield et al. 2021; Tipper et al. 2010). Particles

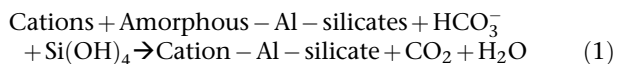present in seawater can partially dissolve or release adsorbed ions (Buck et al. 2010; Jeandel and Oelkers 2015), and their presence can also induce precipitation of supersaturated minerals such as calcite and aragonite where the particles serve as crystallization nuclei (Wurgaft et al. 2016, 2021). Hence, the dual role of particles in seawater as a source and a sink of major elements, complicates the understanding of dissolved major element dynamics.

Studies of the geochemical budgets of major elements often struggle with equating the oceanic sources and sinks of some elements. For example, the current Sr input fluxes are at least a factor of three higher than the hydrothermal and carbonate sinks (Davis, Bickle, and Teagle 2003; Vance, Teagle, and Foster 2009). Similarly, a factor of two or more imbalances between known input and output fluxes were observed for Ca (Milliman 1993; Tipper et al. 2010), Mg (Shalev et al. 2019) and Li (Steiner, Landing, et al. 2022). These imbalances suggest that our current view of which processes are important in the marine cycles of major elements is incomplete. A possible explanation for the observed discrepancies in the marine budgets of alkalinity and major elements is miscounting of silicate mineral dissolution and precipitation. It was demonstrated that dissolution of particulate material carried by rivers to the ocean can equate the isotopic budgets of several elements, including Sr (Jeandel and Oelkers 2015; Jeandel 2016). The counter reaction of aluminosilicate mineral dissolution is clay mineral authigenesis, a process also termed reverse-weathering. Reverse weathering is a potential sink for alkalinity and major elements, and has the following general stoichiometry (Mackenzie and Garrels 1966; Steiner, Rae, et al. 2022):

$$\text{Cations} + \text{Amorphous} - \text{Al} - \text{silicates} + \text{HCO}_3^-$$
$$+ \text{Si(OH)}_4 \rightarrow \text{Cation} - \text{Al} - \text{silicate} + \text{CO}_2 + \text{H}_2\text{O} \quad (1)$$

Biological productivity in the surface ocean, and in particular skeletons formed by marine organisms, consume large quantities of Ca, Sr, and fluorine (F) (de Villiers 1999; Milliman 1974; Rude and Aller 1991), and smaller quantities of other major elements relative to their seawater concentration, including Mg (Morse, Andersson, and Mackenzie 2006) and Li (Langer et al. 2020; Thibon et al. 2021). Sinking of biogenic particles exports shallow water column chemical constituents to the deep ocean (Bishop et al. 1977; Bishop and Wood 2008; Honjo et al. 2008). Major elements thus provide powerful tools to quantify aspects of the biological carbon pump owing to the tendency of different groups of marine organisms to precipitate skeletons with characteristic compositions (Bolden et al. 2023; Milliman 1974; Steiner et al. 2014). Major elements are also instrumental to the ability to reconstruct past ocean environments (Turchyn and DePaolo 2019).

Despite the key role of major elements in the biological carbon pump, few studies have attempted to directly quantify their temporal variability in the oceans (Griffin et al. 2022; Khare et al. 2023; Steiner et al. 2018). To address this gap, we sampled seawater regularly at the northern Gulf of Aqaba, Red Sea, over the course of 2 yr, as part of the *Red Sea Dust, Marine Particulates and*

Seawater Time Series (REDMAST) GEOTRACES process study *GLpr09* (Torfstein et al. 2020). The aim of the current study was to unravel whether there is measurable temporal variability in major element concentrations, and resolve the causes of variability when identified. A main objective of this effort was to explore the extent to which abrupt events such as dust storms and flash floods are important in moderating the chemical composition of the ocean.

Study site

The Gulf of Aqaba is a long (200 km), narrow (maximum width of 16 km), and deep basin (maximum depth of 1800 m) located at the northern tip of the Red Sea (Fig. 1). The Red Sea is connected to the Gulf of Aden and western Indian Ocean 1800 km south of the Gulf of Aqaba via the narrow 140-m-deep Straits of Bab el Mandeb. The Gulf of Aqaba is, in turn, connected

Fig. 1. Location map of the (**a**) Red Sea region and (**b**) north Gulf of Aqaba time-series site at *Station A* on aerial footage from Google Earth. Total suspended atmospheric particles (TSP) are the total suspended atmospheric particles monitoring stations, Interuniversity Institute (IUI) is the on-shore research station. Floodwaters entering the Gulf of Aqaba through the Kinnet Canal and Wadi Shahamon were sampled as part of this study.

to the Red Sea via the 250-m-deep Straits of Tiran. The entire Red Sea—Gulf of Aqaba system is situated in a hyper-arid region between the Sahara and Arabian deserts with average annual rainfall of ~ 25 mm yr^{-1}, with no significant rivers flowing into the Red Sea.

The Gulf of Aqaba deep-water formation happens in winter by cooling and sinking of the surface water, which leads to full mixing of the water column every 5–10 yr (Lazar et al. 2008; Wurgaft et al. 2016). The only significant source of surface water to the northern Gulf of Aqaba is northwest Indian Ocean surface water that flows along the length of the Red Sea (Sofianos and Johns 2002, 2003). Low precipitation and very high evaporation rates due to the dry and warm regional climate result in a northward increase in surface water salinity along the Red Sea from ~ 37 to ~ 40.8 (Steiner et al. 2014; Yao et al. 2014). The salinity gradient drives net flow of surface water from the Gulf of Aden into the Red Sea and toward the Gulf of Aqaba; the average surface water transit time from Bab el Mandeb to the north Gulf of Aqaba is on the order of 1 yr (Yao et al. 2014). The rare rain events that occur in this region occasionally produce short lived flash floods that carry limited volumes of water with high sediment loads (Katz et al. 2015).

Uniquely, the Gulf of Aqaba and Red Sea bottom waters are exceptionally warm, at a year-round temperature $\geq 20°C$ (Carlson, Fredj, and Gildor 2014; Metzl et al. 1989). As a result of the high deep-water temperature, calcite and aragonite saturation is maintained at all depths. High water temperature suggests that kinetics of many chemical reactions is enhanced compared to processes taking place at similar depths in the open ocean. Since the Gulf of Aqaba is the terminal location of the Red Sea surface water circulation, and a site of deep-water formation (Plähn et al. 2002), the seawater composition of the Gulf of Aqaba integrates the processes that modify water chemistry along the entire 2000 km flow pathway of the Red Sea basin. Concomitantly, the sporadic nature of external inputs (e.g., dust storms and flash floods) produces well-defined and temporally isolated events that reflect on processes of potential importance in the open ocean. Based on these characteristics, the Gulf of Aqaba is considered a well-established "natural laboratory" for the investigation of processes that affect water chemistry (e.g., Labiosa et al. 2003; Mackey et al. 2011; Meeder et al. 2012).

The preformed composition of Gulf of Aqaba water deviates from North Atlantic surface waters in its low Ca to salinity ratios (Table 1). The reason is that net precipitation of $CaCO_3$ along the Red Sea removes Ca and alkalinity (Baldry et al. 2020; Krumgalz 1982; Steiner et al. 2014). The absolute concentration of all major elements in the Gulf of Aqaba, including Ca, is higher than any open ocean waters due to the high salinity (40.4–40.8; Fig. 2).

Materials and methods

Sampling

The sampling site (*Station A*, 29°28′N 34°55′E; Fig. 1) is a long-term time-series station of the Israeli National Monitoring

Table 1. Major element ratios in the Gulf of Aqaba ($n = 129$, 1σ SD) compared to surface water samples from the Atlantic Ocean (IAPSO standard collected at $\sim 40°N$ 45–46°W) and Gulf of Aden. Element ratios in Gulf of Aden surface waters are the average of two water samples reported in Steiner, Landing, et al. (2022) and Steiner et al. (2018), the error reported for the two Gulf of Aden samples is the SD of the two samples averaged. Kinnet Canal and Shahamon flood waters (Fig. 1) were collected in the immediate offshore plume of the floods on February 24, 2018.

	Year	Li/Na	Ca/Na	K/Na	Mg/Na	Sr/Na	Sr/Ca	Mg/Ca	Li/Ca
		µmol mol⁻¹	mmol mol⁻¹	mmol mol⁻¹	mmol mol⁻¹	µmol mol⁻¹	mmol mol⁻¹	mol mol⁻¹	mmol mol⁻¹
Gulf of Aqaba average	2017–2018	56.21 ± 0.17	21.706 ± 0.022	21.782 ± 0.026	113.32 ± 0.09	186.93 ± 0.28	8.612 ± 0.009	5.221 ± 0.004	2.589 ± 0.007
IAPSO P157		56.12	21.824	21.781	113.27	186.54	8.547	5.190	2.571
Gulf of Aden surface	Dec 2015, Mar 2018	56.12 ± 0.02	21.812 ± 0.017	21.805 ± 0.030	113.37 ± 0.02	187.34 ± 0.13	8.589	5.198	2.573
Kinnet Canal flood	Feb 2018	214	101.4	22.35	146	1234	12.2	1.44	2.11
Shahamon flood	Feb 2018	56.7	22.14	21.78	113	189.3	8.55	5.12	2.56

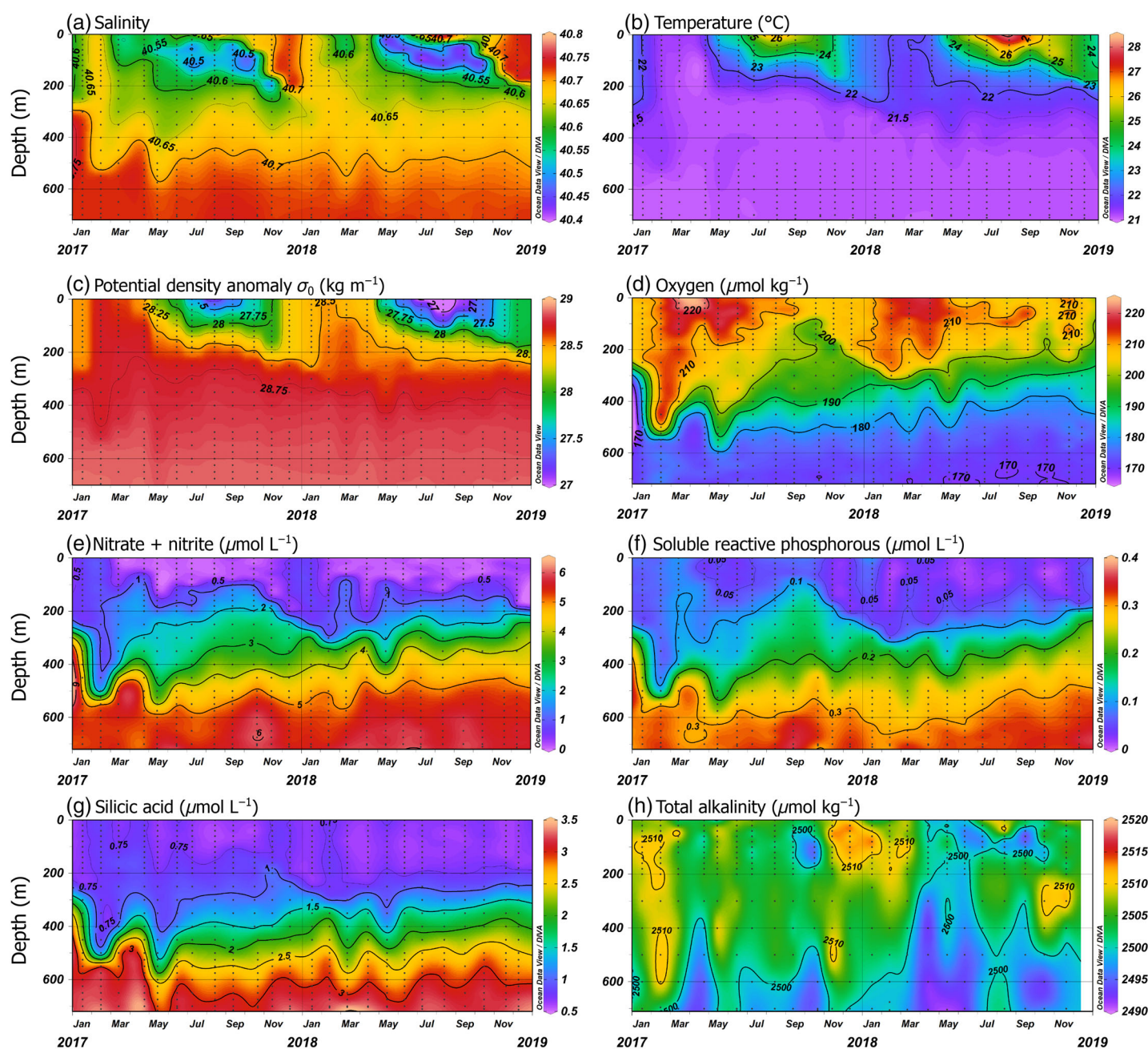**Fig. 2.** Time series of physical and chemical parameters collected at *Station A* in the northern Gulf of Aqaba (data from the NMP; https://iui-eilat.ac.il/Research/NMPMeteoData.aspx).

Program (NMP) at a water depth of ∼ 700 m, visited monthly since 2003. Sampling efforts focused around periods of elevated aerosol fluxes, which in some cases also encompass rain and flash flood events (Supporting Information Fig. S1). The results reported here are from samples that correspond with previously reported dissolved lead concentrations and isotopic compositions (Benaltabet, Lapid, and Torfstein 2020), dissolved aluminum (Al) and silicic acid (Si) (Benaltabet, Lapid, and Torfstein 2022), and (PO_4), manganese (Mn), cobalt (Co), nickel (Ni), zinc (Zn),

and cadmium (Cd) concentrations (Benaltabet, Lapid, and Torfstein 2023); the fully detailed sampling procedures are described there. Briefly, deep seawater profiles were sampled from the RV Sam Rothberg, and shallow profiles (four depths, up to 220 m) were sampled from a fiberglass speedboat. Open seawater samples were collected using acid-cleaned Teflon-coated Go-Flo bottles (General Oceanics), following GEOTRACES sampling protocols (Cutter et al. 2017) and post flood coastal surface seawater samples were collected with acid-cleaned high-density

ployethylene bottles. Sampling bottles were transported within $\sim$ 1–2 h of collection to the Interuniversity Institute for Marine Sciences on shore clean laboratory (Fig. 1), where the seawater was filtered through a 0.2-μm pore size acid-cleaned cartridge filter (Pall AcroPak 1000, Supor Membrane) into acid-cleaned low-density polyethylene cubitainers (10 liters, Fisher Scientific) or bottles (1 liter, Nalgene). Samples were acidified before storage using double distilled 6 N HCl (final concentration 0.024 N HCl) to pH < 1.8. Samples were aliquoted into polypropylene (Corning) tubes for the analyses of dissolved Ca, Sr, K, Mg, and Li concentrations.

Concentrations of total suspended atmospheric particles (TSP) were retrieved from the Israel Ministry of Environmental Protection meteorological monitoring stations, positioned proximally to *Station A* (Fig. 1). Total suspended atmospheric particles concentrations were measured using a Thermo Scientific FH62-C14 continuous particulate monitor (*see* details in Torfstein and Kienast 2018).

Chemical analyses

Calcium, Sr, K, Mg, and Li were analyzed as their ratio to Na by sample-standard bracketing using an Agilent Technology Varian-720 ICP-OES (Inductively Coupled Plasma Optical Emission Spectroscopy) at GEOMAR Helmholtz Centre for Ocean Research Kiel, using the method of Schrag (1999) modified by Steiner et al. (2018). Calibration standards and the consistency standard were different dilutions of an IAPSO salinity standard from batch P157. The concentrations of Sr and Ca in IAPSO P157 were previously measured by isotope dilution and standard addition (Steiner et al. 2018), and the concentration of Li was determined by standard addition (Steiner, Landing, et al. 2022). Our analyses of open ocean water samples from the Southern, Indian, Atlantic and Pacific oceans suggest that there is no measurable variability in K/Na and Mg/Na, hence we assume that previously published K and Mg (Burton 1996) are representative of the IAPSO standard. All samples and consistency standards were diluted in 0.12 M HCl to the same final salinity of 0.425 to match their concentrations (de Villiers, Greaves, and Elderfield 2002; Schrag 1999). Sample analyses consisted of five spectra readings for 10 s each, with each sample analyzed at least one more time after recalibration of the instrument. Conversion from element-to-Na ratios to element concentration assumes Na concentration of 468.46 mmol kg^{-1} at salinity 35, and constant Na-to-salinity ratios, an assumption embedded in the definition of practical salinity (Millero et al. 2008). All major element data presented in this manuscript are normalized to practical salinity of 40.66, which was the average salinity measured during the monthly NMP cruises in the Gulf of Aqaba during 2017–2018. The salinity normalization was designed to eliminate the effect of evaporation on the absolute concentration of major elements, as salinity exerts the main control on major element concentration in the ocean and masks other sources of variability.

Uncertainties were evaluated based on duplicate processing and analyses. Similar uncertainty values were calculated from duplicates analyzed 1 yr apart, suggesting negligible shifts in major element concentrations during storage. The 2σ SD calculated from these long-term evaluations ($n = 30$) are Li 0.10 μmol kg^{-1}, Ca 20 μmol kg^{-1}, K 23 μmol kg^{-1}, Mg 80 μmol kg^{-1}, and Sr 0.16 μmol kg^{-1}.

Oxygen, salinity, alkalinity, and nutrients were analyzed by NMP Eilat using the standardized methods used in the Gulf of Aqaba monitoring program (Shaked and Genin 2018). Oxygen was determined by the Winkler titration method using an automated Titrino 702 SM (Metrohm). Salinity was measured using Micro-Salinometer MS-310e (RBR), an IAPSO salinity standard was used for calibration. Alkalinity was determined by automated titrations of duplicate filtered seawater samples with 0.05 N HCl, and calculated by the Gran titration method. Accuracy of the alkalinity titrations was verified using certified reference material provided by Prof. Andrew Dickson (Scripps Institute of Oceanography). Nitrate plus nitrite, soluble reactive phosphorous (SRP) and silicic acid concentrations were analyzed spectrophotometrically using QuikChem 8000 flow injection analyzer (Lachat Instruments).

Results

Figure 2 presents the NMP time series for the years 2017–2018. The 2 years differed in the depth of winter mixing, which was $\sim$ 500 m in 2017 but < 300 m in 2018. The winter mixing depth affects transfer of nutrients to the surface mixed layer, and consequently the spring phytoplankton bloom (Genin, Lazar, and Brenner 1995; Lazar et al. 2008; Meeder 2012). Winter mixing also bears importance for the long-term nutrient cycle in the Gulf of Aqaba because it ventilates the deep water and enhances export of nutrients from the Gulf of Aqaba to the Red Sea; the same should be of importance for the water column profiles of major elements. In addition, a salinity minimum develops below the surface mixed layer in spring and summer (Fig. 2a). The origin of the salinity minimum is sinking of lower salinity Red Sea surface water flowing into the Gulf of Aqaba in early spring under warmer Red Sea surface water with higher salinity in late spring (Biton and Gildor 2011). The salinity minimum erodes in late autumn when the surface layer cools, promoting deepening of the surface mixed layer (Biton and Gildor 2011). During the study period, the salinity minimum was more pronounced in 2018 than 2017 (Fig. 2a).

Salinity-normalized Ca concentration data suggest that the northern Gulf of Aqaba Ca inventory through the entire water column was slightly lower in 2018 than 2017 (Fig. 3a). A *t*-test confirms this observation, suggesting that the mean Ca concentration in 2018 was $7 \pm 4 \mu$mol kg^{-1} (95% interval, $p < 0.001$) lower than the mean concentration in 2017. The difference grows to $9 \pm 4 \mu$mol kg^{-1} ($p < 0.001$) if only the period of
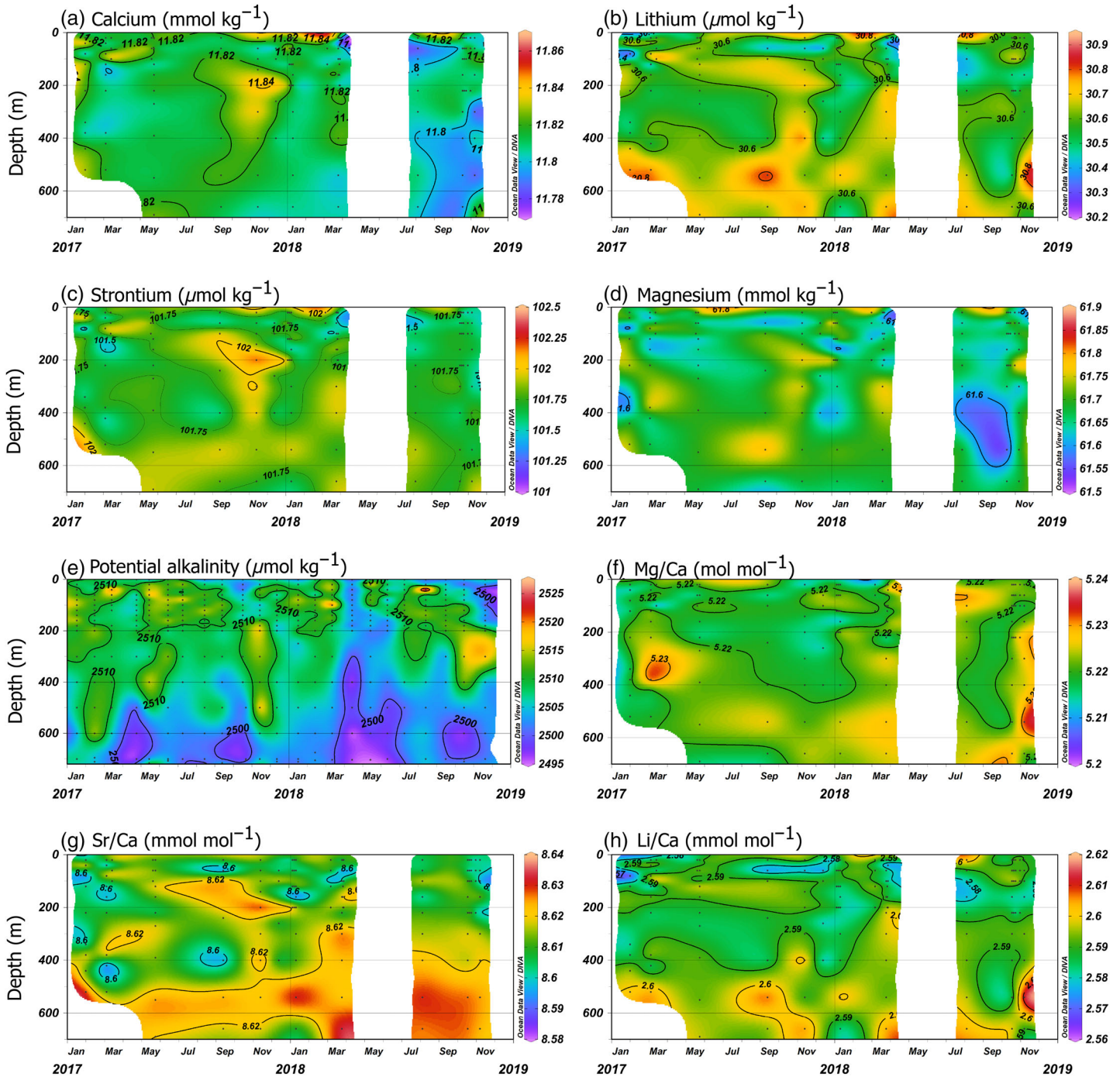
Fig. 3. Time series of salinity-normalized major element concentrations at *Station A* in the northern Gulf of Aqaba.

March–November is considered. If the decline in Ca concentrations results from changes in the carbonate cycle, we expect alkalinity (A_T) to change at a 2:1 ratio with respect to Ca given that precipitation of $CaCO_3$ consumes two charge equivalents per mol of Ca:

$$Ca^{2+} + 2HCO_3^{2-} \leftrightarrow CaCO_3 + CO_2 + H_2O \qquad (2)$$
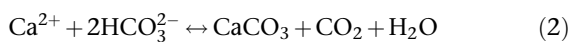
Calculation of $CaCO_3$ precipitation using the A_T data requires normalization of the A_T data to a constant salinity (40.66 in our case) and correction for release of nitrite, nitrate, SRP, and sulfate (Wolf-Gladrow et al. 2007). The nutrient corrected, salinity-normalized A_T is termed potential alkalinity (PA) (Brewer et al. 1975; Steiner et al. 2021).

$$\text{Potential alkalinity} = \frac{\left(A_\text{T} + 1.3 \cdot (\text{NO}_3^- + \text{NO}_2^-) + \text{SRP}\right) \cdot 40.66}{S}$$

(3)

S is the measured practical salinity.

A *t*-test suggests that the 2018 PA was $2.1 \pm 1\ \mu\text{mol kg}^{-1}$ lower than the 2017 PA ($p < 0.001$). The slightly lower concentrations of Ca and PA in the deep water compared to the surface water (Fig. 3a,e) are a general feature of the Gulf of Aqaba and Red Sea (Wurgaft et al. 2016).

Discussion

Carbonate cycle

The decrease in Ca and PA during 2017–2018 reflects increased calcification rates, either along the Red Sea waters on their flow path from the Bab-El-Mandeb Straits to the Gulf of Aqaba, or within the Gulf of Aqaba itself. The long-term A_T trend of the Red Sea suggests that calcification rates were stable between 1977 and 1998 but basin scale calcification rates calculated based on A_T depletion have been lower than past values during 2007–2018 (Baldry et al. 2020; Steiner et al. 2014, 2018), an observation supported by decreased calcification rates measured in central Red Sea coral colonies (Cantin et al. 2010). However, there is no indication for change in Red Sea A_T during 2017–2018 (Baldry et al. 2020; Steiner et al. 2018). Despite the decrease in Gulf of Aqaba PA in 2018, surface water PA was still 25–$30\ \mu\text{mol kg}^{-1}$ higher than 1998 values; this difference is consistent with the increase in alkalinity of the incoming Red Sea water (Steiner et al. 2018) and suggests that net calcification rates in the Gulf of Aqaba were similar in 1998 and 2018, and slightly lower in 2017.

Non-conservative variability in seawater major cation dissolved concentrations can be used to quantify the precipitation/dissolution fluxes of different carbonate and sulfate minerals (Bolden et al. 2023; Steiner et al. 2020). Distinction can be made between calcite, aragonite and celestine (SrSO$_4$ precipitated by Acantharia) based on the distribution coefficient of strontium (D_Sr) into these minerals. A distribution coefficient is the ratio of trace to major element in a mineral to the ratio between the same trace and major element in the solution from which the mineral had precipitated:

$$D_\text{Sr} = \frac{(\text{Sr}/\text{Ca})_\text{mineral}}{(\text{Sr}/\text{Ca})_\text{seawater}}$$

(4)

Generally, calcites are relatively depleted in strontium and have average $D_\text{Sr} \sim 0.17$, aragonites are slightly higher in Sr/Ca than the ambient seawater ($D_\text{Sr} > 1$), and celestine is Sr replete and low in Ca ($D_\text{Sr} >> 1$). The most important exception to this rule is that pteropod aragonite is lower in Sr than most biogenic calcites (Milliman 1974). Common biogenic carbonates can therefore be simplified to a two end-member system

where pelagic carbonates have an average $D_\text{Sr} = 0.17 \pm 0.03$ while corals precipitate aragonite with an average $D_\text{Sr} = 1.04 \pm 0.03$ (Correge 2006; Enmar et al. 2000; Steiner et al. 2014). High abundance of coccolithophorids or organisms precipitating Mg-calcite would yield an intermediate D_Sr of ~ 0.3 (Carpenter and Lohmann 1992; Müller et al. 2014).

Regression of the full Ca and Sr dataset yields a slope of 0.0089 ($R^2 = 0.50$) for material removed/added from Gulf of Aqaba seawater (Fig. 4a). Given the average Gulf of Aqaba Sr/Ca of $0.0086\ \text{mol mol}^{-1}$, the D_Sr is $0.0089/0.0086 = 1.03$, suggesting coral dominance of the local carbonate cycle. The dimensions of the Gulf of Aqaba can explain the importance of benthic over pelagic calcification in this region, but the partition coefficient also suggests that the contribution of benthic calcifiers other than corals is small in the Gulf of Aqaba.

It is reasonable to assume that there is a difference between carbonate reactions in the shallow and deep waters. We thus divide the dataset to samples collected from the top 200 m and samples collected from 400 m to the bottom (Fig. 4a). This division suggests a $D_\text{Sr} = 1.21 \pm 0.09$ in the top 200 m, and 0.81 ± 0.23 from 400 m to the bottom (slope divided by seawater Sr/Ca). The slightly higher than expected D_Sr of the shallow waters is within the range expected for inorganic aragonites (AlKhatib and Eisenhauer 2017), and only slightly higher than coral aragonite D_Sr (Correge 2006; Enmar et al. 2000). The deep-water value is higher than expected because corals do not dominate the carbonate cycle below 400 m. It is possible that inorganic precipitation of carbonate minerals is quantitatively an important process in the supersaturated deep water of the Gulf of Aqaba (Steiner et al. 2019; Wurgaft et al. 2016), this option simultaneously explains the low alkalinity and high D_Sr of the deep water. Another possibility that needs to be considered is that formation and export of Acantharia shells increase the Sr/Ca ratios of the deep water. In the open ocean there is normally a strong positive correlation between Sr and dissolved PO$_4$ concentrations because of the shallow remineralization of Acantharia (de Villiers 1999). The correlation between Sr and PO$_4$ in our dataset is very poor (Fig. 4c), suggesting that in the Gulf of Aqaba Acantharia shells dissolve in the surface waters, likely due to the high water temperature.

A caveat remains in the comparison between the Ca and alkalinity data. Precipitation of CaCO$_3$ consumes Ca and PA at a 1:2 ratio, hence a $7 \pm 4\ \mu\text{mol kg}^{-1}$ decrease in Ca between 2017 and 2018 due to increased CaCO$_3$ precipitation rates should translate to a $14 \pm 8\ \mu\text{mol kg}^{-1}$ decrease in PA, yet the measured decrease in PA was $2.1 \pm 1\ \mu\text{mol kg}^{-1}$. These values were calculated using a *t*-test for the cluster of all samples from each year; though the deviation between the averages is smaller than the single sample uncertainty, the statistical test indicates that they are different ($p < 0.001$). This suggests another sink for Ca or a source for anions. Previous attempts to compare Ca and PA data showed the opposite, with overall excess Ca over PA in the Red Sea, possibly due to dissolution of Messinian gypsum (Steiner et al. 2021). A partial explanation for the discrepancy between the Ca and PA
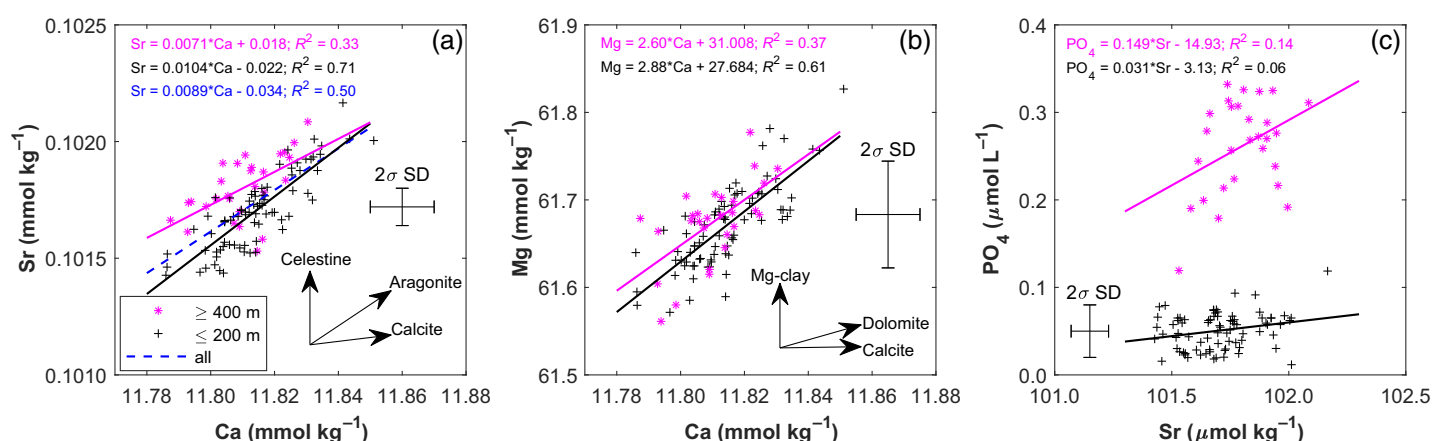
Fig. 4. Linear regressions between Gulf of Aqaba salinity-normalized concentrations of (**a**) calcium and strontium, (**b**) calcium and magnesium, (**c**) strontium and soluble reactive phosphate. The arrows in (**a**) and (**b**) illustrate the slope expected as a result of precipitation/dissolution of common minerals. Note that pteropod aragonite has a lower Sr content than most biogenic calcites, hence the aragonite vector refers to aragonite precipitated by benthic organisms.

in our dataset is that they were not collected at the same time; alkalinity was sampled monthly on NMP cruises while Ca samples were collected at an irregular distribution. Yet, as will be discussed in the following sections, there are indications for dissolution and authigenic precipitation of aluminosilicate minerals in the Gulf of Aqaba; aluminosilicate reactions deviate in their Ca-PA stoichiometry from $CaCO_3$ reactions and may cause the observed discrepancy (Eqs. 1, 2).

Magnesium

Variability in Mg concentrations was previously reported in the Red Sea water mass of the Indian Ocean (Sen Gupta and Naqvi 1984) and in Red Sea surface waters collected in 1998 (Steiner et al. 2014) but was not detected in Red Sea surface water samples collected during 2015–2018 (Steiner et al. 2018).

Calcite shells of benthic organisms can contain up to 20% Mg (Morse, Andersson, and Mackenzie 2006). Assuming seawater Mg/Ca = 5.2 mol mol^{-1}, the Mg distribution coefficient (D_{Mg}) in Mg-calcite is lower than 0.04. The distribution coefficients of Mg in aragonite and pelagic calcites are much lower, around 0.001 (Milliman 1974). The linear fit between the Gulf of Aqaba Mg and Ca data has a slope of 2.68 ± 0.25 mol mol^{-1} or $D_{Mg} = 0.52 \pm 0.05$, with no difference between shallow and deep waters (Fig. 4b). It was suggested, based on Mg isotope measurements, that dolomite ($CaMg[CO_3]_2$) precipitation is far more prevalent in the modern ocean than previously thought (Shalev et al. 2019), and the Gulf of Aqaba would be a reasonable dolomite sink site owing to the high salinity and temperature conditions. There is also a relatively strong positive correlation ($R^2 = 0.64$) between Mg and Ca concentrations in sediment porewater in the Gulf of Aqaba; porewater concentrations of both elements decrease with depth in the sediment, suggesting a common sedimentary sink (James et al. 2021). However, precipitation of dolomite should consume Ca and

Mg at a 1 : 1 M ratio, and the fit we calculate suggests more Mg is reacting.

The only former survey, to our knowledge, that included high quality profiles of dissolved Mg in the Red Sea was conducted in February 1982 and covered 12 stations from the northern Gulf of Aqaba to 25.7°N in the north Red Sea (Krumgalz and Erez 1984). Results of that survey revealed increases in Mg concentrations by up to 2% (1.3 mmol kg^{-1}) from the surface down to 1000 m between 27°N and 28°N, just south of the Strait of Tiran, with high Mg water penetrating halfway through the Gulf of Aqaba. Mg concentrations at the northernmost Gulf of Aqaba station of Krumgalz and Erez (1984), at adjacent location to our station, had slightly lower salinity-normalized Mg concentrations than normal seawater and our measurements. This suggests a considerable sink for Mg in the Gulf of Aqaba. The residence time of water in the entire Red Sea is $\sim$ 35–40 yr (Cember 1988), and it takes between 12 and 40 d for surface water to flow from the Strait of Tiran to the north Gulf of Aqaba study site (Biton and Gildor 2011). Hence, we assume full replacement of water since the Krumgalz and Erez 1982 survey, but the underlying processes that contribute and remove Mg should persist. Steiner et al. (2014) speculated that the northward decrease in Mg concentrations along the Red Sea surface could result from mixing of seawater with Mg poor water from the hot brines that occur on the Red Sea floor. The brine hypothesis is consistent with an observed excess of dissolved Ca over alkalinity in the Red Sea (Steiner et al. 2021), and a small hot brine lake in the Gulf of Aqaba at water depth of 1770 m (Purkis et al. 2022). However, the brine hypothesis only suggests a sink for Mg and is inconsistent with the reverse estuarine circulation of the Gulf of Aqaba because contributions to the deep water would normally flow to the Red Sea and will thus not reach the study site.

The outcrops of Precambrian shield and Cretaceous carbonate formations surrounding the Gulf of Aqaba, provide the weathering products that release Mg, Li, Ca, Sr, and Si to the water in excess of Na, as indicated by the chemistry of flood water entering the northern Gulf of Aqaba (Table 1). Settling Saharan dust further provides reactive minerals and adsorption sites (Torfstein et al. 2017). Circulation of seawater in the rocks and sediments, and dissolution of Messinian evaporites seem to be important factors in the formation of hot brines that dot the Red Sea bottom (Craig 1966; Pierret et al. 2001; Zierenberg and Shanks 1986) but it is not clear from our data if this process is of importance in the cycling of major elements other than Ca. A major sink for the excess supply of some elements is precipitation of various carbonates. Indeed, benthic aragonite precipitated by corals is quantitatively the most important carbonate mineral in this region ("carbonate cycle" section). Excess removal of Mg over Ca suggests authigenic formation of clay minerals may also be important in the Gulf of Aqaba. Indeed, in the following section we show that Li data are consistent with recrystallization of aluminosilicate minerals.

Lithium

Lithium is recognized as a proxy for the inorganic silica cycle because it can readily replace Al, Mg, and iron (Fe) in the crystal lattice of aluminosilicate minerals but has very low partition coefficients into biogenic silica, $CaCO_3$, and organic matter (Langer et al. 2020; Martin, Kastner, and Elderfield 1991; Thibon et al. 2021). Coupled with its low molecular weight, which entails a relatively large difference in mass between the stable isotopes ^6Li and ^7Li, lithium has grown to be a popular proxy of terrestrial weathering and authigenic precipitation of secondary minerals (e.g., Misra and Froelich 2012; Pogge von Strandmann et al. 2012; Tomascak, Magna, and Dohmen 2016). Lithium concentrations in the open ocean vary by up to 3%, with the highest Li concentrations measured in surface waters down-wind of anthropogenic sources (Fabricand, Imbimbo, and Brey 1967; Steiner, Landing, et al. 2022; Stoffyn-Egli and Mackenzie 1984).

Surface water Li concentrations in the Gulf of Aqaba (Fig. 3b) vary but are generally slightly elevated compared to the Gulf of Aden and North Atlantic surface water (Table 1). Many of the profiles record a Li concentration minimum shortly below the surface mixed layer, and Li concentrations are slightly elevated in the deep water (Fig. 5a). Elevated Li concentrations in the surface water and a concentration minimum shortly below the surface mixed layer is a feature observed in several stations from the North Pacific and tropical Indian Oceans (Steiner, Landing, et al. 2022). It was interpreted to reflect the release of aerosol-derived Li at the surface, followed by scavenging of the excess Li by fresh Al and Mn oxides, which are derived from release of Al and Mn from the aerosols. Unlike the deep water of the Gulf of Aqaba, North Pacific deep water is depleted in Li. The difference between the Gulf of Aqaba and North Pacific is consistent with the conclusion of
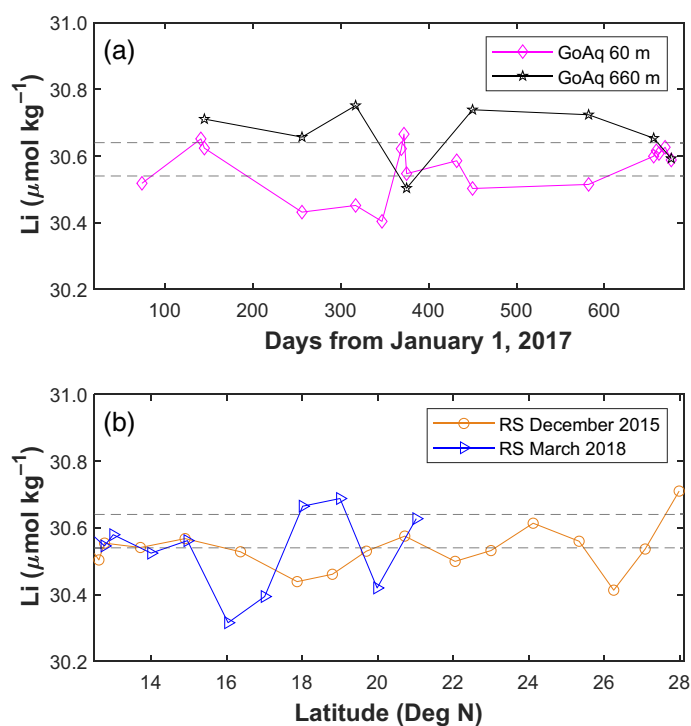

Fig. 5. Time series of lithium concentrations, normalized to Gulf of Aqaba salinity. The distance between the horizontal dashed lines is the analytic 2σ SD. (**a**) Concentrations along equal depth surfaces in the Gulf of Aqaba. (**b**) Spatial transect of lithium concentrations in Red Sea surface waters. Red Sea data are from the Supporting Information of Steiner, Landing, et al. (2022).

Andrews, Pogge von Strandmann, and Fantle (2020) who compiled data from 267 deep-sea drilling sites and found that open-ocean sites tend to be a sink for Li and sites located close to land are more likely to be a source. The reason is that authigenic precipitation of clay minerals outweigh aluminosilicate dissolution in open ocean sediments.

Another indication that seawater Li concentrations vary on short time scales comes from the Red Sea proper (Fig. 5b). On average, Li concentrations did not change along the Red Sea transect, suggesting it is a result of limited supply and removal events that did not last long enough to advect along the basin.

In the upper part of the Gulf of Aqaba water column there is no correlation between the concentrations of Li, Si, and Al (Fig. 6; Benaltabet, Lapid, and Torfstein 2022). Diatoms appear to exert a stronger control on Si concentrations than dissolution of dust in the surface waters (Avrahami, Koplovitz, and Frada 2024; Benaltabet, Lapid, and Torfstein 2022). Despite the continuous supply of dust, we do not observe runaway changes in Li or Al concentrations, as a balance between release and scavenging modulates their concentrations. The lack of correlation between Li and Al, even though they are both tightly linked to the inorganic silica cycle, indicates that the reactions removing them from the dissolved pool might be separated in time and space. Aluminum is quick to react, and its main sink
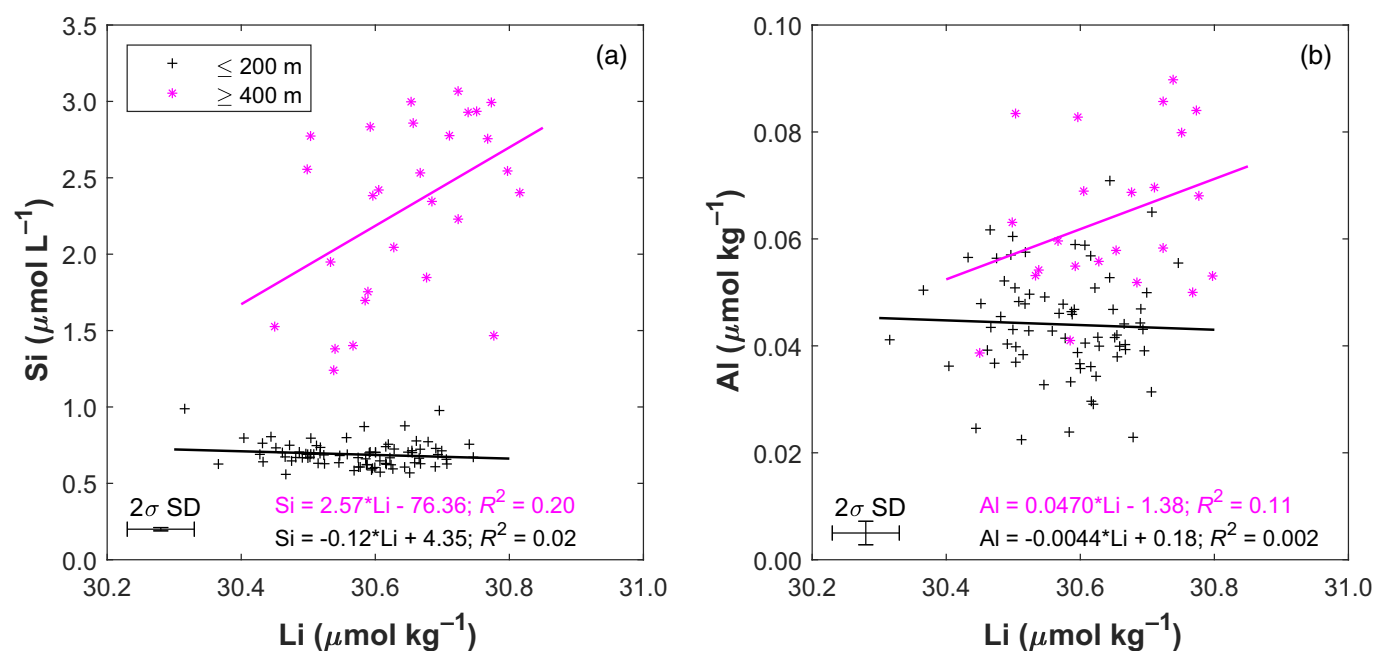
Fig. 6. Linear correlations between salinity-normalized lithium concentration and (**a**) silicic acid, (**b**) aluminum. Aluminum and silicic acid data are from Benaltabet, Lapid, and Torfstein (2022).

in the water column is scavenging onto atmospheric dust particles (Benaltabet, Lapid, and Torfstein 2022). Lithium does not form minerals of its own and is outcompeted in seawater by Na for adsorption onto clay minerals (Hindshaw et al. 2019). Therefore, removal of Li depends on precipitation with Al and Mn oxides (Liu et al. 2015; Takeuchi 1980) or cation replacement in aluminosilicate minerals (Andrews, Pogge von Strandmann, and Fantle 2020). This means a slower reaction rate for Li than Al and a requirement of a-priori formation and/or atmospheric supply of oxides and minerals. In the deeper waters, there is weak positive correlation between Li, Si, and Al, reflecting their common sedimentary source.

Integration

Through calculations of isotope budgets of elements with very different residence times in the ocean, namely strontium, neodymium, silicon, iron, and thorium, Jeandel and Oelkers (2015) demonstrated that release from particulate material has to be a significant input term for these elements to the ocean. The Gulf of Aqaba data support the hypothesis that partial dissolution of externally supplied particulate material is a source of major elements to the ocean; this is particularly clear for the supply of Li from the sediment to the deep water.

Figure 7 summarizes the main pathways that modify major element concentrations in the Gulf of Aqaba. When evaluating the applicability of these findings to open ocean sites it is important to consider some unique characteristics of the study site. The salinity of Gulf of Aqaba water is higher than open ocean sites. The high salinity means high ionic activity

products; hence the saturation states of minerals are higher in the Gulf of Aqaba than under otherwise similar conditions in the open ocean. Studies of the thermodynamic properties of seawater and the carbonate system suggest that at salinity 40 there are still no anomalies in the properties of the solution (Dickson and Millero 1987; Millero and Leung 1976; Mucci 1983). Hence, Gulf of Aqaba salinity still represents seawater conditions and not hypersaline conditions. The surface water temperature at the study site is 21–26°C, which is normal for the tropical and subtropical ocean, but the deep-water temperature is anomalously high. A known implication of the high deep-water temperature is that Red Sea deep water remains supersaturated for aragonite at all depths. However, the high temperature also affects the saturation state and reaction kinetics of aluminosilicate minerals. Dust deposition rates in the Gulf of Aqaba are much higher than the global mean (Benaltabet, Lapid, and Torfstein 2022; Mahowald et al. 2005) and may represent regions like the Sahara plume in the North Atlantic and regions in the Pacific Ocean downwind of East Asian and Australian deserts.

In contrast with trace elements (Benaltabet, Lapid, and Torfstein 2020, 2022, 2023), the short-term effect of aerosol particulate matter supply on major element concentrations in the Gulf of Aqaba is minor. There is no evidence in the new dataset reported here to suggest dust events significantly modify dissolved major element concentrations (Supporting Information Figs. S2–S4). In fact, it seems that the supply of mineral dust acts as a buffer against any large changes in water chemistry. The case is different for the flash flood that occurred on February 24, 2018, where a
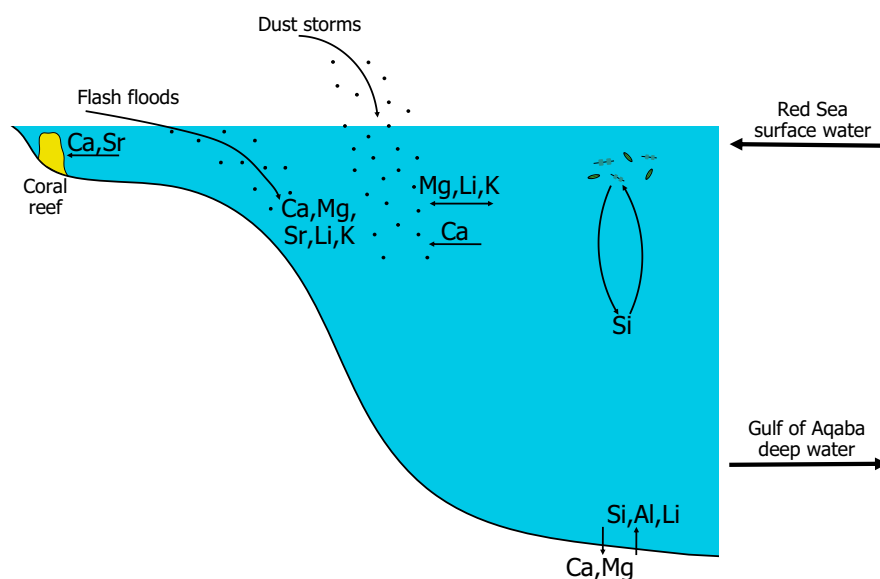
Fig. 7. Schematic diagram of the main sources and sinks of major elements in the northern Gulf of Aqaba. The long-term control of major element concentrations in the Gulf of Aqaba is via water exchange with the Red Sea.

sustained effect of stream carried particular matter on water chemistry was still observed 12 d after the event (Supporting Information Fig. S2).

Magnesium and Li variability imply a very active inorganic silica cycle in the Gulf of Aqaba. Sporadic and significant increases in Li concentrations are most likely the result of desorption and dissolution of incoming aluminosilicates. This is observed both in the surface and deep waters. The surface elevations in Li concentrations are quickly removed, suggesting precipitation into authigenic phases already in the upper water column. Excess uptake of Mg over Ca from the Gulf of Aqaba water is another indication for active precipitation of aluminosilicate minerals, most likely secondary clays.

Evidence for clay authigenesis are available from various ocean regions (e.g., Baldermann et al. 2022; Baronas et al. 2019; Dunlea et al. 2017). The most compelling evidence for clay authigenesis comes from tropical river deltas of the Amazon and Papua New Guinea (Michalopoulos and Aller 1995; Rahman, Aller, and Cochran 2016). Tropical river systems provide large quantities of reactive Al, which are taken in the mineralization process, while diatom frustules serve as nucleation agents and provide the required Si. In the Gulf of Aqaba, high loads of mineral dust and sedimentary inputs provide the Al, Si, and nucleation templates required for clay authigenesis in the absence of diatom frustules in the sediment (Steiner et al. 2019). These observations suggest that precipitation of secondary clays is likely a general case in the ocean, not limited to marginal regions. Note, however, that water temperature may play a key role in the Gulf of Aqaba and tropical rivers given evidence that in low temperature hydrothermal systems Mg uptake rates by the sediment are

low at reaction temperatures $< 15°C$ and increase rapidly as a function of temperature at reaction temperatures $> 15–20°C$ (Shalev et al. 2019).

The observation of an active cycle of aluminosilicate dissolution and precipitation in the water column has far reaching implications for plans to counter ocean acidification using alkalinity enhancement (Gentile et al. 2022; Hartmann et al. 2013) because it implies a strong resistance of the seawater media to variations in its chemical composition (Fuhr et al. 2022). In practice, this means that successful alkalinity enhancement depends not only on dissolving alkaline substances in seawater, but also on avoiding conditions that would trigger precipitation of $CaCO_3$ and authigenic clays, which may counter the effect of the alkalinity enhancement.

Summary

The concentrations of dissolved major elements (Na normalized: Ca, Sr, Mg and Li) are reported in a 2-yr time series from the deep, oligotrophic, hyper-arid Gulf of Aqaba, northern Red Sea, and their main sources and sinks are described. The high-resolution dataset and irregular nature of external inputs allowed the investigation of the chemical interaction between seawater and the boundaries of the Gulf of Aqaba basin. We found that sporadic flash floods are a source of major elements to the Gulf of Aqaba, and that the bottom sediments are a net source of Li, Al, and Si due to dissolution of detrital aluminosilicate minerals.

The variability in Ca and Sr concentrations indicates that the $CaCO_3$ cycle of the Gulf of Aqaba is dominated by changes in coral calcification rates; this is different than

observations from the Red Sea proper where pelagic calcification dominates the $CaCO_3$ cycle.

Variability in Li and Mg concentrations in the Gulf of Aqaba and Red Sea is higher than predicted for biogenic minerals, suggesting an active cycle of aluminosilicate mineral dissolution and formation. The possibility that partial dissolution of aluminosilicate minerals and precipitation of authigenic clays is rapid enough to modify the major element concentrations by a measurable degree, on the time scale of days to months, suggests this may be an important pathway in the biogeochemical cycles of these elements. It is likely that not accounting these fluxes explains at least part of the discrepancy between the known sources and sinks of major elements in the ocean.

Mineral dust deposition does not have a net directional effect on major element concentrations. The reason is that dust particles partially dissolve and release ions to the water, but also form excellent adsorption and nucleation sites. Hence, dust particles may have an active balancing role against large changes in seawater chemistry.

In summary, major element temporal variability in seawater is observed, albeit limited, and is constrained by the interplay between biogenic precipitation rates (e.g., corals), inorganic particle influx from dust and floods along with dissolved terrigenous runoff, as well as interactions with bottom sediments. The results reported here provide a new angle on interpretation of open water major element profiles and their use as monitors of oceanic biogeochemical cycles.

Author Contributions

Zvi Steiner: Conceptualization and funding acquisition; investigation; writing – original draft preparation; writing – review and editing. Tal Benaltabet: Investigation; writing – review and editing. Adi Torfstein: Conceptualization and funding acquisition; investigation; writing – review and editing.

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Supporting Information

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