



Ocean-based Negative Emission Technologies



Deliverable Title	Report on principle kinetic behaviour of selected materials for ocean alkalization purposes
Lead	UHAM
Related Work Package	WP 5
Related Task	Task D5.2
Author(s)	Niels Suijter, Jens Hartmann
Prieto Dissemination Level	Public
Due Submission Date	30.06.2023
Actual Submission	07.07.2023
Project Number	869357
Start Date of Project	01. July 2020
Duration	60 months

Abstract: Examining the dissolution behavior of solid mineral phases for open ocean alkalinity enhancement scenarios with laboratory-based experiments was basically impractical. The general occurrence of secondary mineral precipitation during dissolution experiments led to a shift of focus towards the stability of alkalinity and the description of the observed runaway precipitation processes. Based on detailed examinations of the triggering factors for the observed precipitation, results from the stability experiments derived upper feasible limits for the implementation of open world OAE application scenarios.



This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement number 869357.

Document History

Date	Version	Description	Name/Affiliation
07.07.2023	1.0	e.g.: First submitted version, reviewed and validated by...	Suitner, Hartmann/UHAM

Disclaimer: This document reflects only the author's view and the European Commission and their executive agency are not responsible for any use that may be made of the information it contains.

List of abbreviations, acronyms and definitions

Ber	Bergen
CDR	Carbon Dioxide Removal
d	day(s)
DIC	Dissolved Inorganic Carbon
Exp./E	experiment
Fig.	figure
GC	Gran Canaria
Hel	Heligoland
kgsw	kg seawater
NETs	Negative Emission Technologies
OAE	Ocean Alkalinity Enhancement
S	salinity
TA	Total Alkalinity
Temp.	temperature
$\Omega_{\text{aragonite}}$	saturation state of aragonite

List of tables and figures

Table 1: Overview of conducted stability experiments during field campaigns.

Figure 1: Dunite dissolution experiments

Figure 2: Calcium hydroxide dissolution experiment

Figure 3: Magnesium hydroxide dissolution experiment after 24 hours

Figure 4: Stability experiment, magnesium hydroxide, Gran Canaria

Figure 5: TA development over time, biotic, non-CO₂-equilibrated, Bergen

Figure 6: TA-DIC diagram with stability ranges, biotic, non-CO₂-equilibrated, Bergen

Figure 7: Comparison Gran Canaria – Bergen, non-CO₂-equilibrated

Figure 8: TA development over time, abiotic, CO₂-equilibrated, Bergen

Figure 9: Comparison equilibrated and non-equilibrated experiments, Bergen

Figure 10: Ikaite - stability and solubility in seawater

1. Introduction

1.1 Context

OceanNETs is a European Union project funded by the Commission's Horizon 2020 program under the topic of Negative emissions and land-use based mitigation assessment (LC-CLA-02-2019), coordinated by GEOMAR | Helmholtz Center for Ocean Research Kiel (GEOMAR), Germany.

OceanNETs responds to the societal need to rapidly provide a scientifically rigorous and comprehensive assessment of negative emission technologies (NETs). The project focuses on analyzing and quantifying the environmental, social, and political feasibility and impacts of ocean-based NETs. OceanNETs will close fundamental knowledge gaps on specific ocean-based NETs and provide more in-depth investigations of NETs that have already been suggested to have a high CDR potential, levels of sustainability, or potential co-benefits. It will identify to what extent, and how, ocean-based NETs can play a role in keeping climate change within the limits set by the Paris Agreement.

1.2 Purpose and scope of the deliverable

Our contribution to the assessment of Ocean Alkalinity Enhancement (OAE) involved conducting abiotic laboratory experiments during the mesocosm campaigns in Gran Canaria 2021 (WP5.3), Bergen 2022 (WP5.4) and Helgoland 2023. While contributing to the impact assessments of OAE during these campaigns, we covered various processes related to the deployment of alkalinity addition scenarios.

According to the tasks of WP5.1 the aim is to examine the behaviour of particles undergoing dissolution, changes in the solid surface because of seawater interaction, determine the stability of the alkalinity and if secondary phases might precipitate during the dissolution process. Findings from the dissolution experiments revealed that the characterization of dissolution kinetics and stability of minerals with high potential for ocean alkalization approaches could not be determined. Using solid mineral powders to generate alkalinity in seawater, as initially planned, resulted in intense secondary mineral formation during experiments, removing previously created alkalinity out of the system and leading in some cases to a net-loss of TA, below the initial seawater TA level. Latter would be counteracting the idea of OAE and limiting its potential. Besides the work of Moras et al. (2022) we described this process of so called "runaway precipitation" in our recent publication (Hartmann et al., 2023). Two additional publications are going to include insights from the campaigns in Bergen (Suitner et al. (a), in prep.) and Heligoland (Suitner et al. (b), in prep.) describing temporal and chemical ranges for a safe and effective application of OAE and the effect of dilution processes to prevent the runaway process.

Besides observing the effects of the runaway precipitation process during our laboratory-based experiments, the same procedure was also observed during the mesocosm studies on Gran Canaria and Helgoland, underlining the importance to understand this process in detail to secure safe and sustainable application scenarios. Adjusting to these findings

our research primarily focused on the above-mentioned tasks to describe the stability of alkalinity and the process of secondary mineral formation.

1.3 Relation to other deliverables

The experiments in WP 5.1 delivered additional insights to the abiotic carbonate system response during the field campaigns on Gran Canaria 2021 (WP5.3) and in Bergen 2022 (WP5.4). By excluding the influence of organisms from the system, we could isolate abiotic and biotic driven processes. In close cooperation with various partners from WP5.3/4 we contributed to the approach to provide an assessment on the suitability of various OAE strategies, different types of minerals, their application for ocean alkalization purposes and providing limiting factors for open world applications. Data sets and results from these experiments are available for all project members.

2. Results

2.1 Dissolution experiments

During lab-based dissolution experiments in seawater, various solid materials (Ca-, Mg-hydroxides, olivine-rich dunite, basalt, limestone) were tested for their potential for OAE. Apart from basalt, all dissolution processes were affected by secondary carbonate formation, removing the previously gained alkalinity and by that hinder the determination of dissolution rates. Dependent on the type of material, these precipitation processes could arise in seconds to days after the dissolution process was started.

2.1.1. Olivine-rich dunite

In case of dunite 500–700 $\mu\text{mol/kgsw}$ alkalinity were generated after 1 day, using application rates of 3.3 g/kgsw. Nevertheless, first signs of secondary Ca-carbonate formation, indicating the potential start of a runaway process, could be observed after 4 days (see Fig. 1), similar to precipitation processes occurring in benthic system described by Fuhr et al. (2022). Within the first hour, dissolution rates ($1.0\text{--}3.7 \cdot 10^{-8}$ mol olivine $\text{s}^{-1}\text{m}^{-2}$, at 23 °C and an average grainsize of 50 μm) were comparable to computed olivine dissolution rates, using calculation schemes based on Hangx & Spiers (2009) and Köhler et al. (2013), but were declining continuously afterwards. After 24 hours the dissolution process basically stopped after dissolving around 0.9% of the added mass. Assuming an open ocean application, material would constantly be in contact with fresh undersaturated seawater, which could sustain dissolution rates, but nonetheless the bulk of the material would sink to the ocean floor, significantly reducing its CO_2 sequestering efficiency and questioning its applicability. In addition, applying materials with slow dissolution kinetics provide surface area for precipitation, lowering the energy barrier to form secondary phases, potentially resulting in the observed runaway processes.

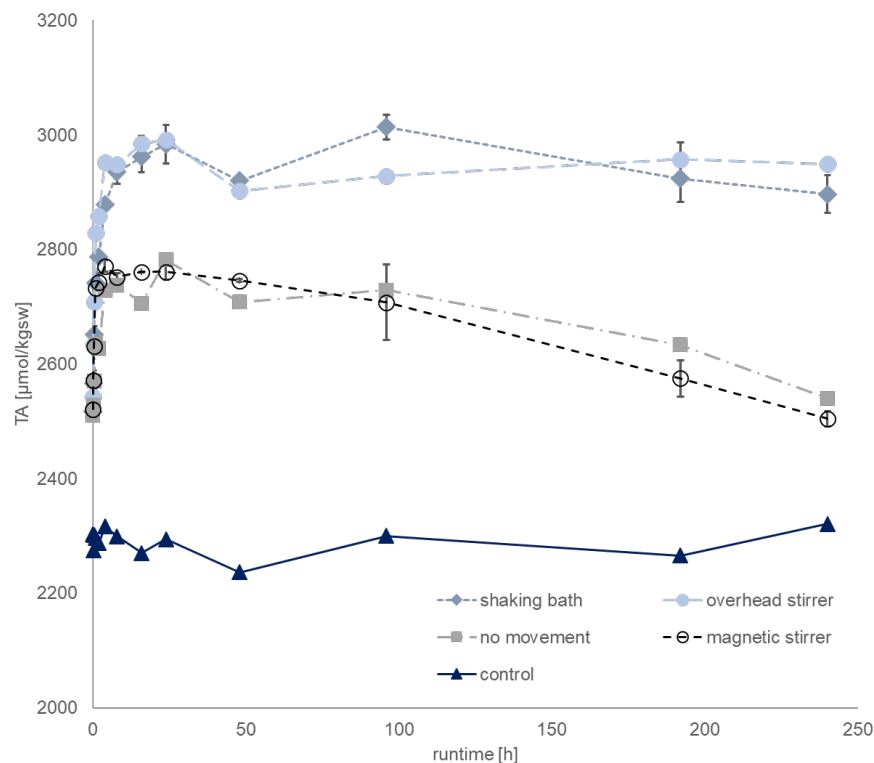


Figure 1. Development of TA over time during dunite dissolution experiments, using different shaking/stirring methods

2.1.2 Calcium hydroxide

As CaO and MgO would transform fast to hydroxides a focus was set on the hydroxides. Using solid Ca-hydroxides for alkalinity enhancement in our experiments led in some cases to complete dissolution of the added material within the first 10 seconds. Figure 2 shows the development of TA, pH and $\Omega_{\text{aragonite}}$ during the dissolution of 0.66g of calcium hydroxide per kgsw over a runtime of 10 days. Although the added mass was below the solubility of calcium hydroxide in seawater, no TA could be generated during this process. On the contrary, instead of a gain of TA, a substantial net-loss of alkalinity of about 1500 $\mu\text{mol/kgsw}$ (initial seawater TA: 2297 $\mu\text{mol/kgsw}$), due to intense secondary mineral formation after several minutes, was observed. Multiple forward and reverse reactions during the runtime of this experiment, reflected by pH and TA variations, were indicating alternating dissolution and precipitation procedures of secondary phases, showing that at least parts of precipitates are re-dissolvable (see Hartmann et al. (2023) for details). The rapid dissolution occurring concurrently with the prevalence of parallel precipitation rendered the determination of dissolution kinetic rates in these experiments unfeasible and not target-oriented.

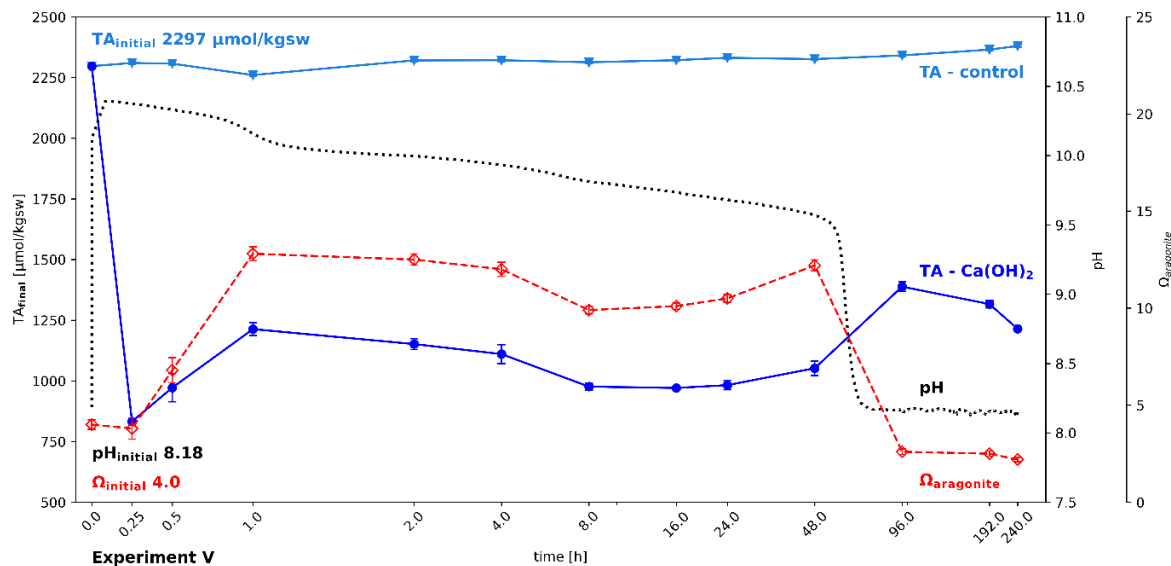


Figure 2. Partial recovery of alkalinity after carbonate precipitation; change in alkalinity, pH, and $\Omega_{\text{aragonite}}$ after adding 0.66 g $\text{Ca}(\text{OH})_2/\text{kgsw}$ to North Sea water in a shaking bath (simulating wave processes to allow accelerated air–water– CO_2 exchange). $\text{Ca}(\text{OH})_2$ dissolution is so fast that it is hard to track. While the water gradually equilibrates with the atmosphere, a series of loss and gain of alkalinity can be observed (see Fig. 7 in Hartmann et al. (2023) for details).

2.1.3 Magnesium hydroxide

To showcase that the dissolution of hydroxides in seawater could generate additional alkalinity, figure 3 showcases the use of varying masses of magnesium hydroxide. After 24 hours of runtime, treatments above 0.066g $\text{Mg}(\text{OH})_2/\text{kgsw}$ exhibited a net-loss of TA due to precipitation. The dissolution of smaller amounts yielded an alkalinity enhancement up to 1600 $\mu\text{mol}/\text{kgsw}$, close to the ideal potential of the added masses. Nevertheless, this increase was not permanent as one of the follow-up experiments indicated (Fig. 4), also see Hartmann et al. (2023), Fig. 5.

In cooperation with our project partners from GEOMAR during the Gran Canaria field campaign, alkalinity was increased by using magnesium hydroxide. Two different $\text{Mg}(\text{OH})_2$ materials were tested in abiotic (0.2 μm -filtered seawater– excluding biota) and biotic (50 μm -filtered seawater – including phytoplankton) setups in parallel over 4 days. Except for the treatment with an TA increase by 600 $\mu\text{mol}/\text{kgsw}$ (equivalent to 0.019g $\text{Mg}(\text{OH})_2$) all other treatments once again experienced a net-loss of alkalinity after 4 days, showing that the achieved TA increase at day 0, measured 5 min after start of the experiment, was not sustainable.

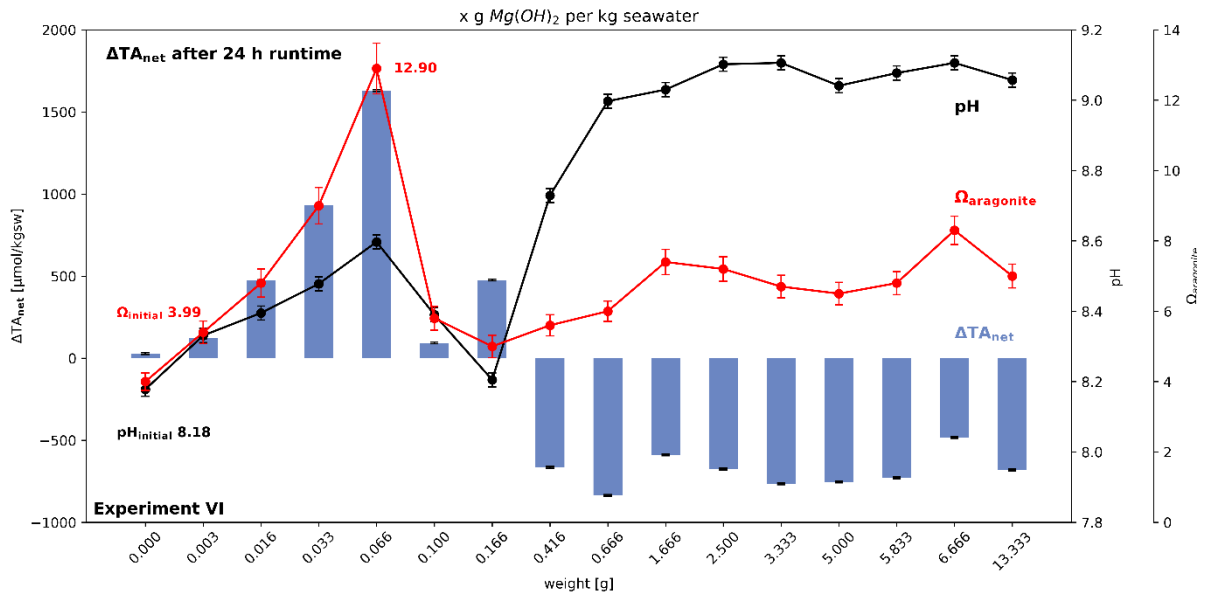


Figure 3. Change in alkalinity (TA_{net}), pH and $\Omega_{aragonite}$ after adding $Mg(OH)_2$ to North Sea water in a shaking bath (simulating wave processes to allow accelerated air–water– CO_2 exchange). Net loss occurs when alkalinity addition exceeds the critical saturation level of aragonite (see Fig. 8 in Hartmann et al. (2023) for details).

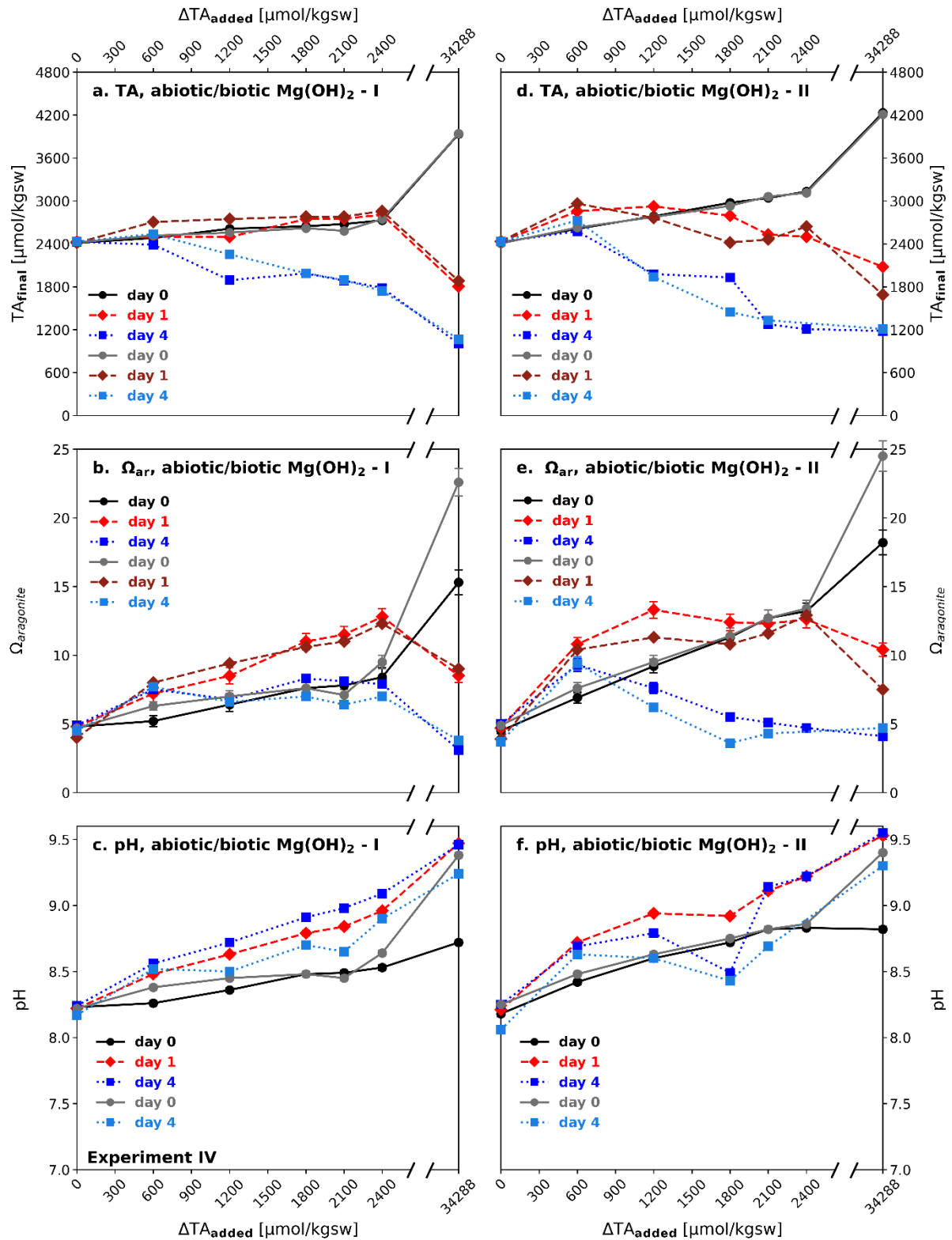


Figure 4. Dissolution of particulate $Mg(OH)_2$ (types I and II) over time: (a, d) development of alkalinity (TA_{final}); (b, e) aragonite saturation state ($\Omega_{aragonite}$); and (c, f) pH immediately after addition (day 0 – black/grey), 1 d (red), and 4 d after (blue); abiotic treatments – black, red, and dark blue graphs; biotic – grey and light blue graphs. Brucite additions range from 19.2 mg (ΔTA_{600}) to 1000 mg (ΔTA_{34288}). Comparable trends are observed in all treatments, independent of substrate type (I and II) and set-up (abiotic/biotic). Figure from Hartmann et al. (2023).

2.1.4 Summary dissolution kinetics

In summary, determining the dissolution kinetics of the proposed materials in the context of OAE was impractical. Dissolution rates of silicates were too low to consider these substances for open ocean application, while materials like oxides or hydroxides dissolve within seconds after addition and the omnipresence of precipitation processes basically was prohibiting any kind of determination of reliable dissolution rates in seawater. In context of potential future implementation of solid-phase alkalinity enhancement, our findings indicate that it is advisable to limit the extent of TA to moderate levels (max. 300–600 $\mu\text{mol/kgsw}$), while using fast dissolving mineral phases, until suitable distribution techniques inhibiting significant losses of alkalinity due precipitation of carbonates are identified. Any kind of provided surface, like the added materials itself, might trigger heterogenous precipitation. These processes could be rapid and microenvironments around a dissolving particle might reach overcritical values almost in an instant.

2.2 Stability of alkalinity experiments

Building on our experiences from the dissolution experiments, we increasingly focused on the stability of alkalinity after the enhancement. The objective was to identify limits of alkalinity enhancement in case alkalinity can be supplied to seawater, for example with prepared solution, like from reactors. Through the utilization of NaOH or $\text{Na}_2\text{CO}_3/\text{NaHCO}_3$ stock solutions to introduce the alkalinity, potential uncertainties associated with employing solid phases were eliminated (due to the fast dissolution) and alkalinity levels of the used seawater could be increased with high precision. In cooperation with our partners from GEOMAR diverse incubation experiments were conducted during field campaigns on Gran Canaria 2021, Bergen 2022 and Helgoland 2023, to examine relevant factors determining the stability of alkalinity of TA enhanced seawater and describing the characteristics of related secondary mineral formation processes. To give an overview, Table 1 lists all carried out experiments, including brief descriptions with supplementary information.

To test the durability of the added alkalinity, series of gradient approaches were conducted, increasing TA in a non- CO_2 -equilibrated (NaOH) and a CO_2 -equilibrated way (mixture of $\text{Na}_2\text{CO}_3/\text{NaHCO}_3$). Equilibrated alkalinity addition simulated scenarios wherein the CO_2 uptake of the ocean was already achieved, meaning that in addition to an elevation in TA, DIC was adjusted in a way that pCO_2 values are in an equilibrium to an atmosphere of 420ppm. The enhancement employing NaOH in the non- CO_2 -equilibrated approaches simulated only an addition of TA, before an equilibration with the atmosphere is achieved. For both options stability experiments were carried out in different environments, using local natural seawater from Gran Canaria (S 36, Temp. 23°C, runtime 4 days), Bergen (S 32.5, Temp. 10–14°C, runtime 20–25 days) and Helgoland (S 30, Temp. 8–12°C, runtime 20 days). By enhancing the TA levels by more than $\approx 2400 \mu\text{mol/kgsw}$ in a non- CO_2 -equilibrated manner an immediate decline in TA values could be observed. Treatments above this level uniformly showed a fall back to a specific salinity and temperature dependent TA level, indicating immediate formation of secondary phases. Treatments with an increase below 2400 $\mu\text{mol/kgsw}$ progressively

started losing alkalinity over the runtime of the experiments. To give an impression, Fig. 5 shows the development of TA over time in the biotic non-CO₂-equilibrated experiments in Bergen.

Table 1 Overview of conducted stability experiments during field campaigns. Data owner abiotic: UHAM (Niels Suitner, Jens Hartmann), biotic (GC): GEOMAR (Jan Taucher, Carl Lim), biotic Bergen (Ber): GEOMAR (Giulia Faucher, Carl Lim) and Ber E6 (Julieta Schneider), Helgoland (Hel): (Niels Suitner (UHAM), Giulia Faucher (GEOMAR)). If not mentioned, all experiments were conducted with abiotic and biotic treatments.

Location/ Exp. #	Description
GC E1	CO ₂ -equilibrated, runtime: 4 days, lab-experiment, material used: Na ₂ CO ₃ /NaHCO ₃
GC E2	Non-CO ₂ -equilibrated runtime: 4 days, lab-experiment, material used: NaOH
GC E3	Non-CO ₂ -equilibrated, runtime: 4 days, lab-experiment, material used: Mg(OH) ₂ – I (solid)
GC E4	Non-CO ₂ -equilibrated, runtime: 4 days, lab-experiment, material used: Mg(OH) ₂ – II (solid)
GC E5	CO ₂ -equilibrated, runtime: 10-(160) days, triggering precipitation with solid aragonite, only biotic treatments, harbor of Taliarte, Na ₂ CO ₃ /NaHCO ₃
Ber E1	CO ₂ -equilibrated, runtime: 20 days, incubation box, material used: Na ₂ CO ₃ /NaHCO ₃
Ber E2	Non-CO ₂ -equilibrated runtime: 25 days, incubation box, material used: NaOH
Ber E3	Non-CO ₂ -equilibrated runtime: 4 days, incubation box, comparison mesocosms I, only biotic
Ber E4	Non-CO ₂ -equilibrated runtime: 4 days, incubation box, comparison mesocosms II, only biotic
Ber E5	Non-CO ₂ -equilibrated runtime: 48 hours, incubation box, high resolution, only biotic
Ber E6	CO ₂ -equilibrated, runtime: 9 days, triggering precipitation with solid aragonite, only biotic treatments, incubation box, Na ₂ CO ₃ /NaHCO ₃
Hel E1	Non-CO ₂ -equilibrated runtime: 20 days, incubation box, material used: NaOH, abiotic
Hel E2	Non-CO ₂ -equilibrated runtime: 20 days, incubation box, material used: NaOH, biotic
Hel E3	Non-CO ₂ -equilibrated runtime: 3 days, dilution after changing timesteps, biotic
Hel E4	Non-CO ₂ -equilibrated runtime: 6 days, dilution after changing timesteps, biotic
Hel E5	Non-CO ₂ -equilibrated runtime: 5 days, testing varying dilution intensities, biotic

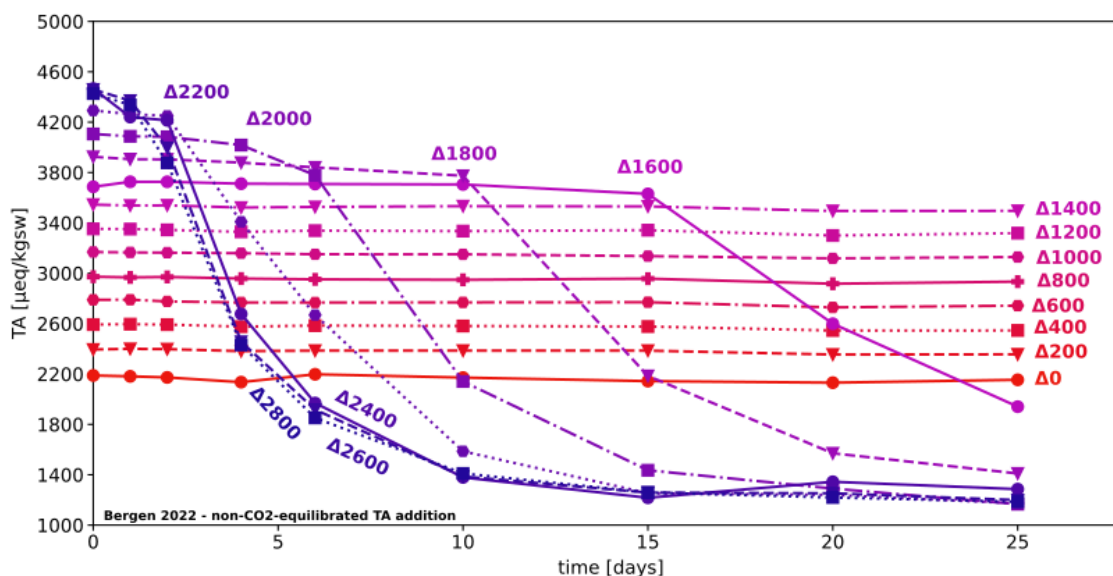


Figure 5. TA development over time, non-CO₂-equilibrated (biotic), showing a sigmoidal decline, due to secondary carbonate mineral precipitation, y-axis absolute measured TA values, treatments are labelled with the initial increase in alkalinity in $\mu\text{mol}/\text{kgsw}$ (e.g. $\Delta 1800$), used seawater $\approx 2200 \mu\text{mol}/\text{kgsw}$ ($\Delta 0$). Notice that without immediate precipitation $\Delta 2600$ and $\Delta 2800$ should have reached 4800 and 5000 $\mu\text{mol}/\text{kgsw}$, figure taken from Suitner et al. (in prep.).

As mentioned above, treatments with an immediate decline uniformly fell back to a TA of $\approx 4500 \mu\text{mol}/\text{kgsw}$, while other treatments showed a delayed start of the decline process. If a decline was observed, treatment entered the runaway precipitation process as described by Moras et al. (2022) and Hartmann et al. (2023). Generated precipitates potentially provided fresh mineral surfaces, which increased the potential for triggering even more precipitation, finally leading to a positive feedback loop. The characteristic sigmoidal decline patterns showed higher decline rates in higher treatment levels and delayed start in treatments between $\Delta 2400$ and $\Delta 1600$. Treatments below $\Delta 1600$ stayed constant over the runtime of 25 days, which does not exclude the possibility of an initiation of the precipitation process if the experiments would have been continued. For all samples which underwent the complete runaway process, it uniformly ended around $1200 \mu\text{mol}/\text{kgsw}$ in TA. Notice that treatments below $\Delta 2400$ hold the potential to prevent the runaway process, by diluting the enhanced water mass. Deduced from this dataset Fig. 6 is showing time and TA-ranges until a treatment with the indicated TA and DIC values should be diluted to avoid negative consequences, due to secondary precipitation. The shown ranges vary, depending on the chemical properties (e.g., TA, DIC, salinity) of the manipulated mass of water and environmental factors like temperature. In comparison to the above-mentioned ranges of $300\text{--}600 \mu\text{mol}/\text{kgsw}$ for the employment of solid substances, here we could show for conditions like in Bergen, an increase above $2000 \mu\text{mol}/\text{kgsw}$ would be possible if sufficient dilution could be ensured. Nevertheless, the observation of an immediate decline above $\Delta 2400$ sets an upper threshold for practical application scenarios, while also setting limits to the efficiency of the whole concept of OAE, as it is not possible to increase the alkalinity to arbitrary levels. Understanding related processes and its guiding influence factors might be crucial

to secure a safe and efficient implementation of OAE in open world systems. Shown ranges in Fig. 6 could vary greatly and should not be generalized, each mass of water needs to be assessed individually to ensure a safe and sustainable application.

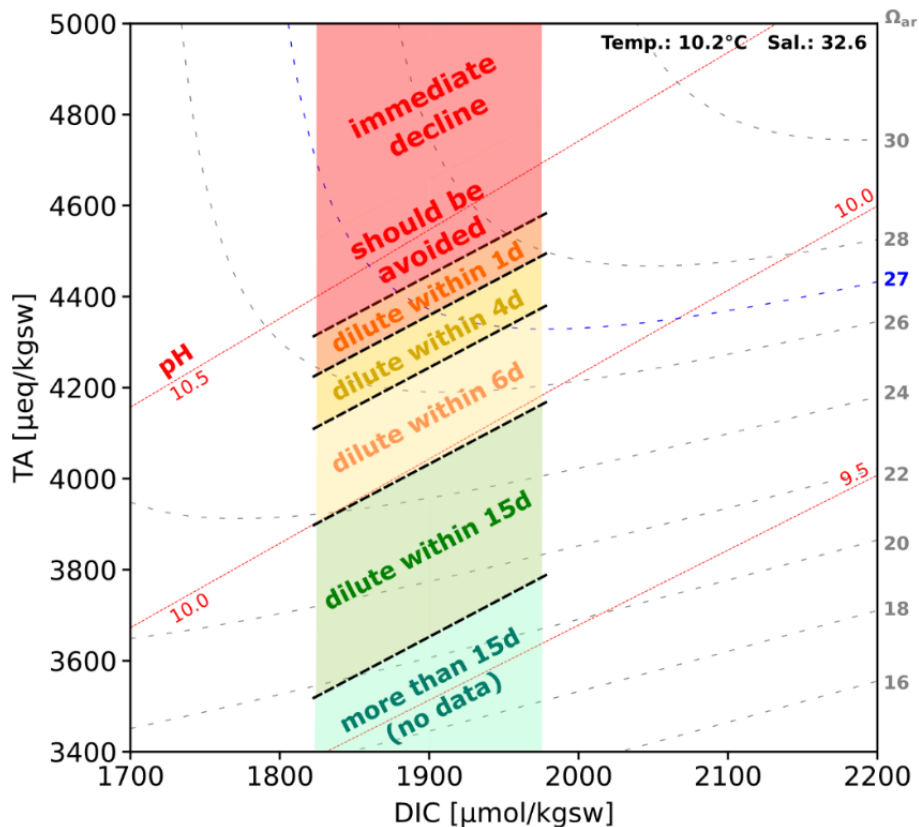


Figure 6. TA-DIC diagram, showing temporal ranges until a TA enhanced water mass should be diluted to avoid secondary mineral precipitation (note: given time for dilution in the figure is location specific and needs to be identified individually). Including contours of pH and saturation states of aragonite. Only valid for TA, DIC, salinity and temperature ranges reached during the Bergen campaign, d: time in days, figure taken from Suitner et al. (in prep.)

To underline the importance of the framework parameters, Fig. 7 is showing the direct comparison of Gran Canarian and Bergen data for the non-CO₂-equilibrated experiments. While in both setting an immediate decline could be observed (green arrows), conditions on Gran Canaria (S 36, temp. 23 °C, TA≈2400 µmol/kgsw, DIC≈2200 µmol/kgsw) were favorable for faster precipitation than in Bergen (S 32.5, temp. 10 °C, TA≈2200 µmol/kgsw, DIC≈1950 µmol/kgsw). While the precipitation process already came to an end after 4 days on Gran Canaria, a comparable treatment (yellow dot) in Bergen stopped the runaway process after 10–15 days.

While the non-CO₂-equilibrated alkalinity enhancement in the Bergen setup allowed an increase around 2200 µmol/kgsw, without immediate precipitation, the CO₂-equilibrated approaches, using the same seawater, reached values above Δ9000 without immediate precipitation (see Fig. 8 and 9). Nonetheless, secondary mineral precipitation was also

triggered during the equilibrated experiments and again treatments began entering the runaway process progressively. After 20 days treatments above $\Delta 3600$ experienced a loss of TA and the precipitation process stopped for treatments above $\Delta 5200$. Here, final TA values after the end of the runaway process aligned on a diagonal baseline, resulting from a constant final $\Omega_{\text{aragonite}}$ (Fig. 9). In general, during all conducted experiments the runaway precipitation stopped if the water reached $\Omega_{\text{aragonite}}$ values between 3–6, comparable to findings in Moras et al. (2022).

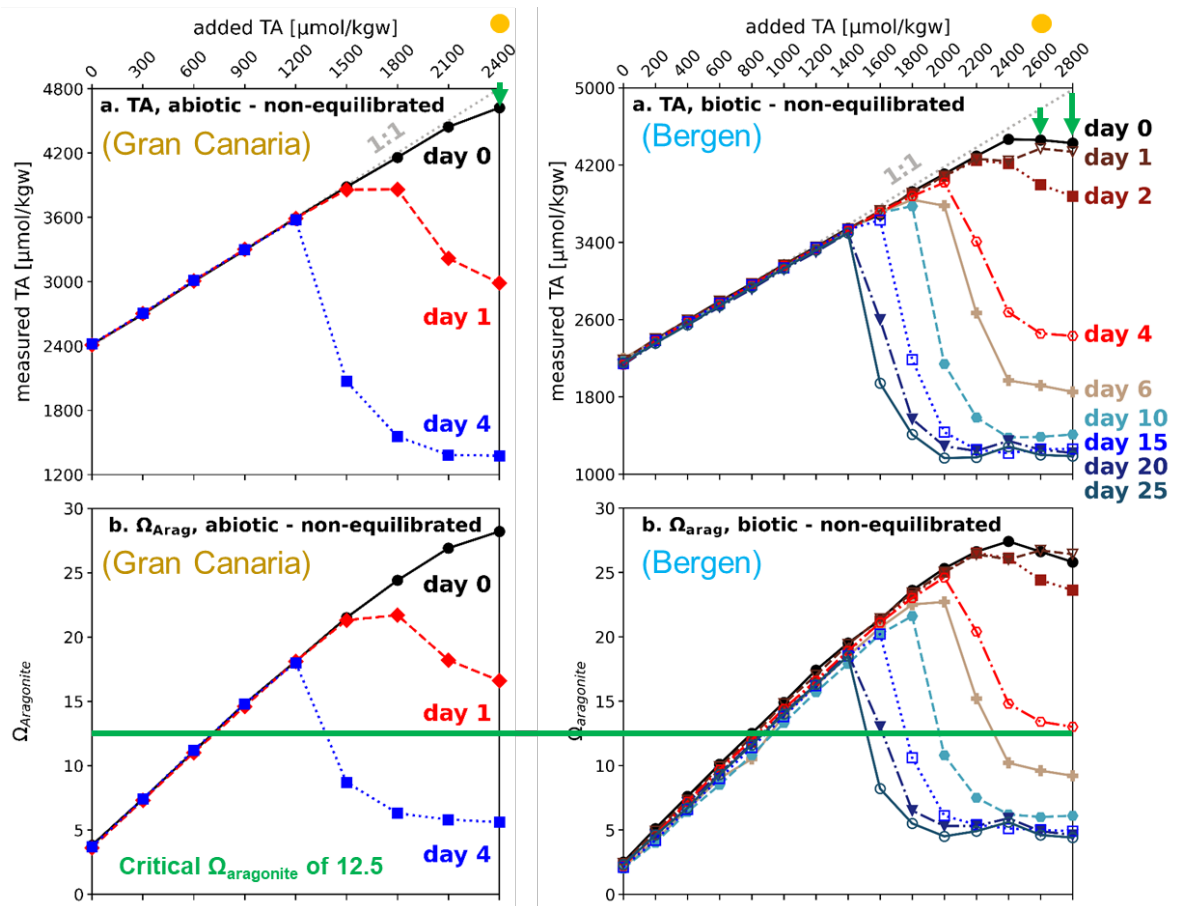


Figure 7. Comparison of TA and saturation state of aragonite development during the non- CO_2 -equilibrated stability experiments on Gran Canaria and Bergen.

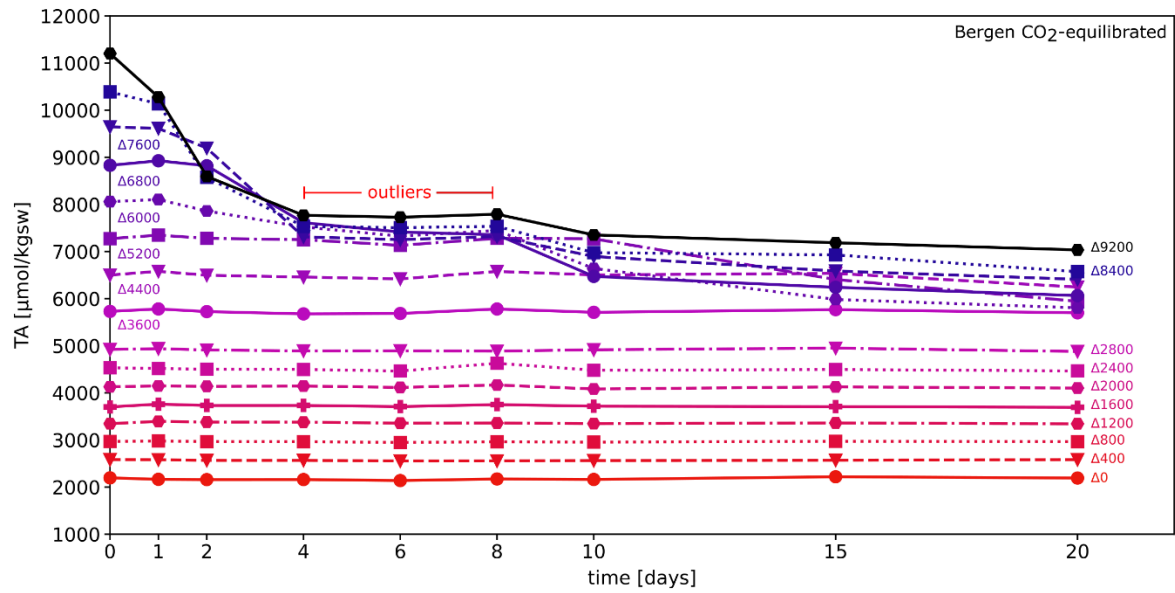


Figure 8. TA development over time, CO₂-equilibrated (abiotic), again showing sigmoidal decline, due to secondary carbonate mineral precipitation, y-axis absolute measured TA values, treatments are labelled with the initial increase in alkalinity in µmol/kgsw (e.g. Δ2800), used seawater ≈ 2200 µmol/kgsw (Δ0). Notice the TA loss of around 4000 µmol/kgsw in the highest treatments after the runaway process ended. Nevertheless, a net-gain in TA was achieved, but accompanied by a net-acidification of the manipulated water, if the runaway process was triggered, see Fig. 9. Figure taken from Suitner et al. (in prep.).

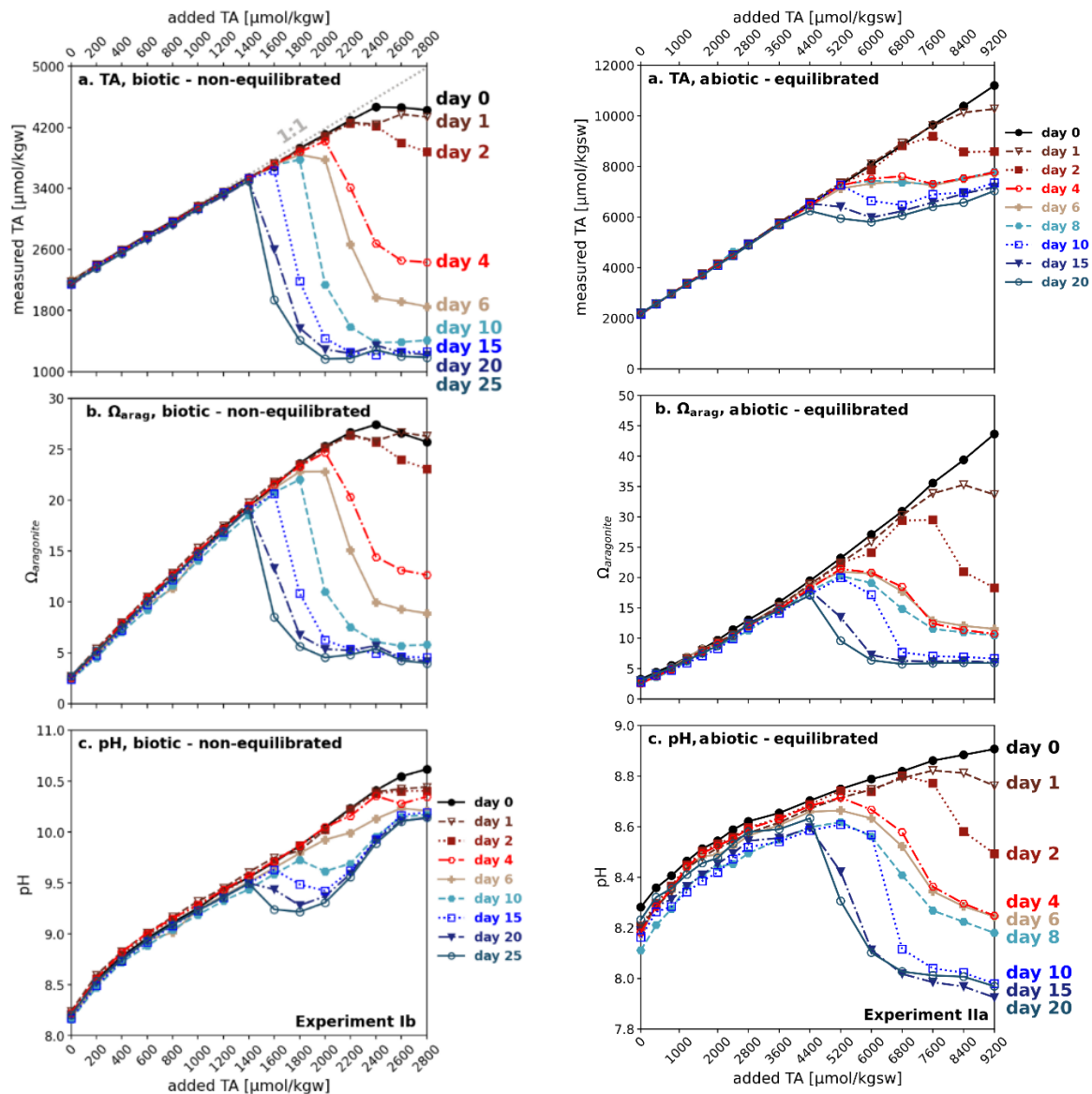


Figure 9. Comparison of TA, pH and saturation state of aragonite in the non-CO₂-equilibrated and equilibrated experiments in Bergen, starting parameters: S 32.5, temp. 10–13 °C, TA ≈ 2200 μmol/kgsw, DIC ≈ 1950 μmol/kgsw, notice the net-loss in TA in the non-CO₂-equilibrated approach (top left) and the net-acidification in the CO₂-equilibrated experiment (bottom right), figure taken from Suitner et al. (in prep.)

2.3 Preliminary data - Helgoland

Early data from the just finished Helgoland campaign showed that dilution could in fact stop or delay the formation of secondary phases, as proposed above in Fig. 6. Different times and degrees of dilution were tested in a comparable setting to the Gran Canaria and Bergen experiments.

2.4 Stability of artificially produced ikaite for OAE application

In a separate experimental setup, the mineral ikaite was artificially produced, a calcium carbonate with six water molecules (CaCO₃·6 H₂O), which in contrast to calcite dissolves

in seawater (Fig. 9 C). The stability of this mineral was tested after creation before it transforms into calcite, which does not dissolve in seawater. It was found that the ikaite can be stored for several days at low temperatures, giving time to transport it to the application area. It was tested if the ikaite dissolves fully in seawater and nearly 100% efficiency was found in the laboratory test (Fig. 9 D). The precipitates are shown in Fig. 9 (A) and (B). This work was published in Renforth et al 2022.

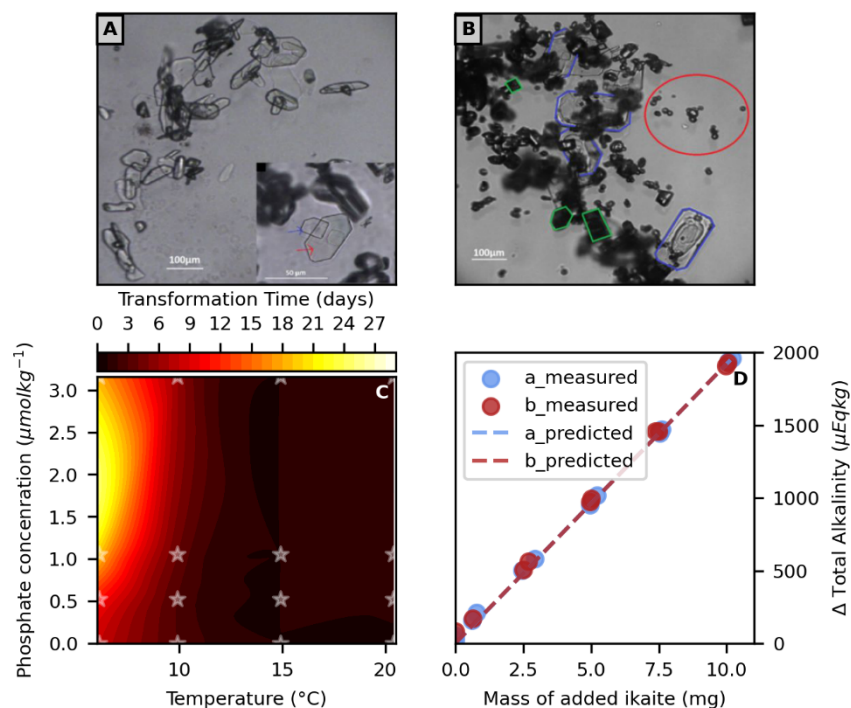


Figure 10. (A and B) Optical characterization of powder that was interspersed with different crystal habits of ikaite showing (A) morphologies that often characterize ikaite crystals 10 and (B) assemblage of ikaite (blue edges), vaterite (red circle), and calcite (green edges). (C) A stability matrix of ikaite showing presence within the solution over time for a given temperature (interpolated between the marked datapoints). (D) Measured and dissolved total alkalinity (TA) from dissolution of ikaite in two natural seawaters (A and B), This figure is from Renforth et al. (2022, Fig. 1).

3 Conclusion

Examining the dissolution behavior of solid mineral phases for open ocean alkalinity enhancement scenarios with laboratory-based experiments was basically impractical, due to the slow or too fast dissolution kinetics of the proposed substances and in particular because of the omnipresent occurrence of secondary carbonate precipitation. The application of solid materials potentially lowered the energy barrier to form secondary phases, by providing surface area for homogenous precipitation. Accordingly, our research focused on the task to describe the stability of alkalinity and processes related to the precipitation effect observed in almost all conducted experiments.

Understanding processes, triggering factors and consequences of the examined runaway precipitation might be crucial for a feasibility assessment of the whole concept of OAE. The observed spontaneous precipitation in our experiments might help to define practical upper TA addition limits for application scenarios. In general, runaway precipitation processes should be avoided in any case, as related consequences would negate the previously added alkalinity. For open world implementations a sufficient dilution of the manipulated water should be ensured around the injection point to avoid negative effects. Besides the direct introduction of substances in the ocean water, various other technical options to provide alkalinity were proposed (e.g. Eisaman et al., 2023), offering possibilities to mitigate or avoid negative effects during the injection process.

4 References

- Eisaman, M., Geilert, S., Renforth, P., Bastianini, L., Campbell, J., Dale, A., ... & Rønning, J. (2023). Chapter 3: Assessing the technical aspects of OAE approaches. *State of the Planet Discussions*, 2023, 1-52.
- Fuhr, M., Geilert, S., Schmidt, M., Liebetrau, V., Vogt, C., Ledwig, B., & Wallmann, K. (2022). Kinetics of olivine weathering in seawater: an experimental study. *Frontiers in Climate*, 39.
- Hangx, S. J., & Spiers, C. J. (2009). Coastal spreading of olivine to control atmospheric CO₂ concentrations: A critical analysis of viability. *International Journal of Greenhouse Gas Control*, 3(6), 757-767.
- Hartmann, J., Suitner, N., Lim, C., Schneider, J., Marín-Samper, L., Arístegui, J., ... & Riebesell, U. (2022). Stability of alkalinity in Ocean Alkalinity Enhancement (OAE) approaches—consequences for durability of CO₂ storage. *Biogeosciences Discussions*, 1-29.
- Köhler, P., Abrams, J. F., Völker, C., Hauck, J., & Wolf-Gladrow, D. A. (2013). Geoengineering impact of open ocean dissolution of olivine on atmospheric CO₂, surface ocean pH and marine biology. *Environmental Research Letters*, 8(1), 014009.
- Moras, C. A., Bach, L. T., Cyronak, T., Joannes-Boyau, R., & Schulz, K. G. (2022). Ocean alkalinity enhancement—avoiding runaway CaCO₃ precipitation during quick and hydrated lime dissolution. *Biogeosciences*, 19(15), 3537-3557.
- Renforth, P., Baltruschat, S., Peterson, K., Mihailova, B. D., & Hartmann, J. (2022). Using ikaite and other hydrated carbonate minerals to increase ocean alkalinity for carbon dioxide removal and environmental remediation. *Joule*, 6(12), 2674-2679.