Reply to comments on EGUSPHERE-2025-381 (30th April 2026)

'Report # 1' (Suggestions for revision or reasons for rejection), Anonymous Referee #1, (1st March 2026)

Authors' responses

Comment 1

I appreciate the authors' efforts in responding to my previous comments and believe the manuscript has improved as a result. That said, I still find it challenging to identify the extent to which this study advances our understanding of runaway precipitation beyond the authors' earlier work. The authors note that additional studies are underway with the goal of developing more general models that incorporate environmental parameters, thereby increasing predictive applicability. In my view, integrating the present manuscript with this broader effort would considerably strengthen the contribution and impact of the work. At the same time, I do not see any fundamental flaws in the study as presented. Therefore, if the editor considers the contribution to be sufficient for the scope and standards of the journal, I support its publication after consideration of my comments below.

Thank you very much for your comments and suggestions. We appreciate the time taken to review our manuscript and the feedback provided. The comments have been carefully considered and addressed in this revision, and we hope that they significantly enhance the manuscript's focus, clarity, and readability. Below you will find a point-by-point reply to each comment of the current revision round (1st March 2026).

Additional minor edits were applied across the manuscript to enhance comprehensibility and readability. Please note that some of the section, figure, and table numbers have changed due to the applied edits.

Regarding the novelty of this work in comparison to our previous study, Suitner et al. (2024) focused on identifying critical thresholds for the stability of alkalinity, with a focus on describing and interpreting the experimental dataset and identifying upper thresholds for TA additions. Although saturation state and surface area were already recognized as the main drivers, no attempt was made to develop a mechanistic, generalized framework to describe it.

In the present study, we aimed to provide such a framework, with the goal of enabling the implementation of the described runaway precipitation patterns and associated processes into modelling approaches. While this study builds on the same dataset and uses the curve-fitting procedure, it extends the previous work by demonstrating that the measured TA evolution can be traced using established theoretical concepts. Within the limitations of relying on a single dataset that is only valid and fine-tuned for the present environmental conditions (also see Comment 8), the assumptions about the key drivers underlying the TA loss process in Suitner et al. (2024) could be confirmed here as a proof of concept.

To clarify and strengthen the novelty, the related paragraph in the Introduction:

Old:

"Suitner et al. (2024) demonstrated the potential of utilizing inverse logistic functions to depict the temporal evolution of the TA-loss process during the runaway carbonate formation phase (see Fig. 1). In this study, principal descriptive parameters such as TA addition and stability ranges to trigger the runaway process or the timespan of the precipitation phase could be formalized based on their experimental dataset. This approach also offers the possibility of a straightforward integration of time-dependent loss terms into predictive computational models simulating OAE addition scenarios, as presented by He & Tyka (2023), Ou et al. (2025), Schwinger et al. (2024) or Zhou et al. (2024)."

was adjusted to:

New: (**L80-88**):

"Suitner et al. (2024) demonstrated the potential of inverse logistic functions to describe the temporal evolution of the TA loss during the runaway carbonate formation phase (see Fig. 1). Building on this concept, this study presents a proof-of-concept mechanistic kinetic framework that combines empirically derived rate equations with measurable parameters (such as carbonate saturation state and particle surface area) to reconstruct the temporal evolution of TA loss and identify the main drivers of the process. The resulting parameterization is currently only calibrated to the experimental dataset and does not yet include additional environmental controls such as temperature, salinity, or suspended particles, but it provides a transferable calculation scheme that could later be implemented in predictive ocean models simulating OAE addition scenarios, as discussed by He & Tyka (2023), Ou et al. (2025), Schwinger et al. (2024), and Zhou et al. (2024)."

Comment 2

Removal of outliers: I am doubtful of this approach. I appreciate that sometimes results deviate without a clear cause, but if no apparent issues were found, I don't see why the data should be removed. Since both treatments with deviating data were filtered, is it possible that the reduced number of particles drives a different precipitation pattern? I would suggest keeping the data, or at least calculating the results both with and without the "outliers" for comparison.

Following the suggestion, more emphasis was placed on describing the handling of outliers in the filtered approaches. A new section (3.2 Outliers) was added to the main text to clarify the circumstances and rationale for removing outliers from the calculations of the empirical rate equations and the curve fitting process. Additionally, a set of diagrams (also see Figs. R1-3 below) and tables was added to the supplements to compare results with and without the outlying data points (curve fits Figs. S1 and S2, and empirical rate equations Fig. S3 and Tabs. 1 and 2).

New section (**L199-216**):

"3.2 Outliers

In the experimental series performed in the Raunefjorden, a systematic cessation of the precipitation process was observed in two of the three experimental setups. Precipitation arrested after 6 days, in the eq filtered approach (Fig. S1) and after 8 days in the filtered neq approach (Fig. S2). For all TA treatments that had already entered the accelerated precipitation phase, no further significant changes were detected in any measured parameter.

Each TA treatment level was distributed to three individual bottles to minimize headspace effects while providing sufficient volume for sampling. The bottles were sampled sequentially three to four times each, after which a fresh set of bottles was opened. Although both affected series were run concurrently, a logistical offset of two days separated their start times. All anomalies in the two independent series occurred between June 24th and 28th 2022, specifically in the second set of opened bottles, while the first and last sets of bottles displayed a regular precipitation pattern. This temporal clustering suggests that an external factor, such as water temperature, sunlight intensity, or a particular aspect of the sampling procedure, may have systematically influenced the affected reactor bottles. Repeated measurements with calibrated standards ruled out an analytical error, and the concurrent impact on both pH and TA confirms the reliability of the recorded values. Consequently, the two sampling days that exhibited these anomalies were excluded from the curve-fitting calculations. A comparison of the fits with and without these data points is shown in supplementary Figures S1, S2, and S3."

<u>**Comment 3**</u>

Throughout the text: Be careful when discussing R. Sometimes it is described as the $CaCO_3$ precipitation rate, and at other times as the TA loss rate. These are not the same (the latter is twice as high as the first).

Thanks a lot for noticing. Wording has been adjusted in several cases.

<u>**Comment 4**</u>

Methods: I still believe more experimental detail is needed. Readers should not have to consult a separate publication to understand the rough experimental design, and it should not be assumed that readers have read the previous paper. The overview of the experimental setup must include all relevant background information (in short form) – section 2.1 should include information on all sample collections, not just the Norwegian gradient experiment. The Gran Canaria sampling comes out of the blue.

Section 2.1 has been reworked and extended to provide additional details regarding the experimental procedures for both the Raunefjorden and the Gran Canarian campaign, detailed descriptions for the experimental setups are provided in the supplements.

<u>Old:</u>

"Three incubation experiments were conducted to examine the stability of TA of the local "filtered" (mesh size 0.2 µm) and "unfiltered" (mesh size 50 µm) seawater of the Raunefjorden, Norway (60.27° N, 5.20° E). Within TA-gradient approaches, 250 ml polystyrene cell culture bottles were filled with natural seawater and incubated in a flow-through incubation box, following the natural light and temperature conditions. Runaway precipitation was observed in eq and neq treatments, after surpassing specific time and TA addition ranges, allowing the description of patterns during the precipitation process. A detailed description of the experimental results, design and methods is already given in Suitner et al. (2024), a brief overview is also provided in Tab. 1 (see supplements for further details)."

New: (**L106-126**)

"All data analyzed in the present study derive from TA-gradient experiments described in Suitner et al. (2024). During a field study from May to July 2022, natural seawater was collected at the Espeland marine station (Raunefjorden, Bergen, Norway; 60.27° N, 5.20° E). Within 20-25 days lasting TA-gradient approaches, 250 ml polystyrene cell culture bottles were filled with "filtered" (mesh size 0.2 µm) or "unfiltered" (mesh size 50 µm) natural seawater ($TA_{initial}$ ~2190 ± 10 µmol kg^{-1}, DIC ~1890 ± 20 µmol kg^{-1}, pH ~8.25 ± 0.05, Ω_{ar} 2.8 ± 0.4 and Sal. ~32.6 ± 0.1) and incubated in a PMMA flow-through incubation box, mimicking the natural light and temperature conditions (10-16°C). Each TA level was distributed among 3-4 bottles to allow sequential sampling while minimizing head-space. TA was either increased with a 0.5 M NaOH (neq) or with a mix of 0.4 M $NaHCO_3$ and 0.2 M Na_2CO_3 (eq) stock solutions to maintain ambient pCO_2 levels (~420 µatm). TA, pH, salinity, conductivity and temperature were measured using a Metrohm 888 Titrando (0.02 M HCl titration) and a WTW MultiLine® multimeter. An overview of the experimental setup is given in Tab. 1; full methodological details are provided in the supplementary information (SI).
The precipitated particles of three selected filters (0.2 µm PC), collected during incubation experiments within previous campaigns published in Suitner et al. (2024) (neq ΔTA_{2600} and ΔTA_{2800}, Raunefjorden) and Hartmann et al. (2023) (neq ΔTA_{2400}, Gran Canaria) were used to analyze particle sizes, morphology and their sinking velocities. Similar to the Raunefjorden, the filters from the Gran Canaria experiment were obtained from a TA-gradient experiment, conducted in local oligotrophic seawater ($TA_{initial}$ ~2411 ± 5 µmol kg^{-1}, DIC ~2006 ± 16 µmol kg^{-1}, pH ~8.15 ± 0.02, Ω_{ar} 4.4 ± 0.3, Sal. ~36.6 Temp. ~23 °C), designed to assess the stability of alkalinization approaches (see SI for further details). Incubations during the Gran Canarian campaign ran for 4 days and followed the same analytical protocol as the Norwegian series introduced above. A detailed description of the Gran Canaria setup is provided in the SI."

Comment 5

L152-157: This info should be in section 2.1.

Thanks a lot for noticing, the basic parameters of the used seawater (Raunefjorden and Gran Canaria) will be provided in section 2.1. Also, see Comment 4.

Comment 6

Section 3.4: Given the circular nature of the correlations between parameter b and the induction time, and between parameter c and the APP (all derived from the same time-series data), I don't understand why a whole section is devoted to discussing these relationships. It should suffice to mention in one sentence in a previous section that the induction time and APP can be derived from the model. Variations in the coefficients and in the induction time/APP timespan will be due to data scatter (measurement precision, slight differences in exactly when the changes in the curve are estimated to begin and end, etc.).

As suggested section 3.4 was deleted and replaced by a short paragraph at the end of section 3.1. The related Fig. 6 was relocated to the supplements.

Old:

"3.4 Prediction of onset and timespan of APP

The established continuous logistic functions allow estimations of effects occurring between measurement points, thereby improving the overall accuracy beyond what discrete experimental datasets could provide. Based on the 40 µmol kg^{-1} d^{-1} TA-loss criterion (see section 3.2 and sketch Fig. 5), these functions could therefore assess the initiation of the APP for specific initial TA and Ω_{ar} levels, as well as a given starting particle surface area (see regressions in Fig. 4). In this regard, Fig. 6 illustrates the correlations of the curve-fitted coefficients (b) and (c) and their related entities of the modeled induction times and APP timespans (see Fig. 2). Under the present physicochemical conditions, the provided regressions could be utilized as conversion equations to estimate the TA development of a treated water mass based on an existing database or to convert observational data into mathematically expressible equations for predicting the future evolution. Note that the high correlations of coefficients (b) and (c) with induction times and APP timespans are relate to the circumstance that both coefficients are determined through the described numerical curve-fitting procedure, which is based on the properties of the logistic function and thus indirectly incorporates information about induction times and APP timespans."

New: (**L194-197**)

"The continuous logistic curve-fits allow the estimation of the APP onset and duration. Parameter **(b)** correlates with the induction time, while **(c)** correlates with the APP timespan (see Figs. 4 and 5). Under the present conditions the regressions can be used as conversion equations for TA evolution. Related conversions and regressions are provided in the SI (Fig. S7)."

Comment 7

Section 3.5: Equation 2 has already been defined above, so this does not need to be repeated.

L274 Redundant definition will be deleted as requested.

Comment 8

Section 4.1: While the authors reply to my comment, they have not made any modifications to the text. It is still stated that the model allows predictions of TA stability ranges (L314-316), which is only true if experiments have been run in exactly the same conditions. The authors also claim that the curve fit model might be integrated into ocean models. Again, this is only useful if the TA stability model is mechanistic and accounts for basic environmental conditions.

To address the concerns and improve clarity, we have added the following two sentences at the end of section 4.1 General findings:

(**L342-345**)

"The relationships reported herein are specific to the experimental setup and the particular environmental conditions under which they were measured. Consequently, projections or models that apply the presented precipitation patterns and equations may not hold when conditions differ from those of the original study."

and additional these statements in the introduction:

(**L81-86**)

“[…], this study presents a proof-of-concept mechanistic kinetic framework that combines empirically derived rate equations with measurable parameters such as carbonate saturation state and particle surface area to reconstruct the temporal evolution of TA loss and identify the main drivers of the process. The resulting parameterization is currently only calibrated to the experimental dataset and does not yet include additional environmental controls such as temperature, salinity, or suspended particles, […]”

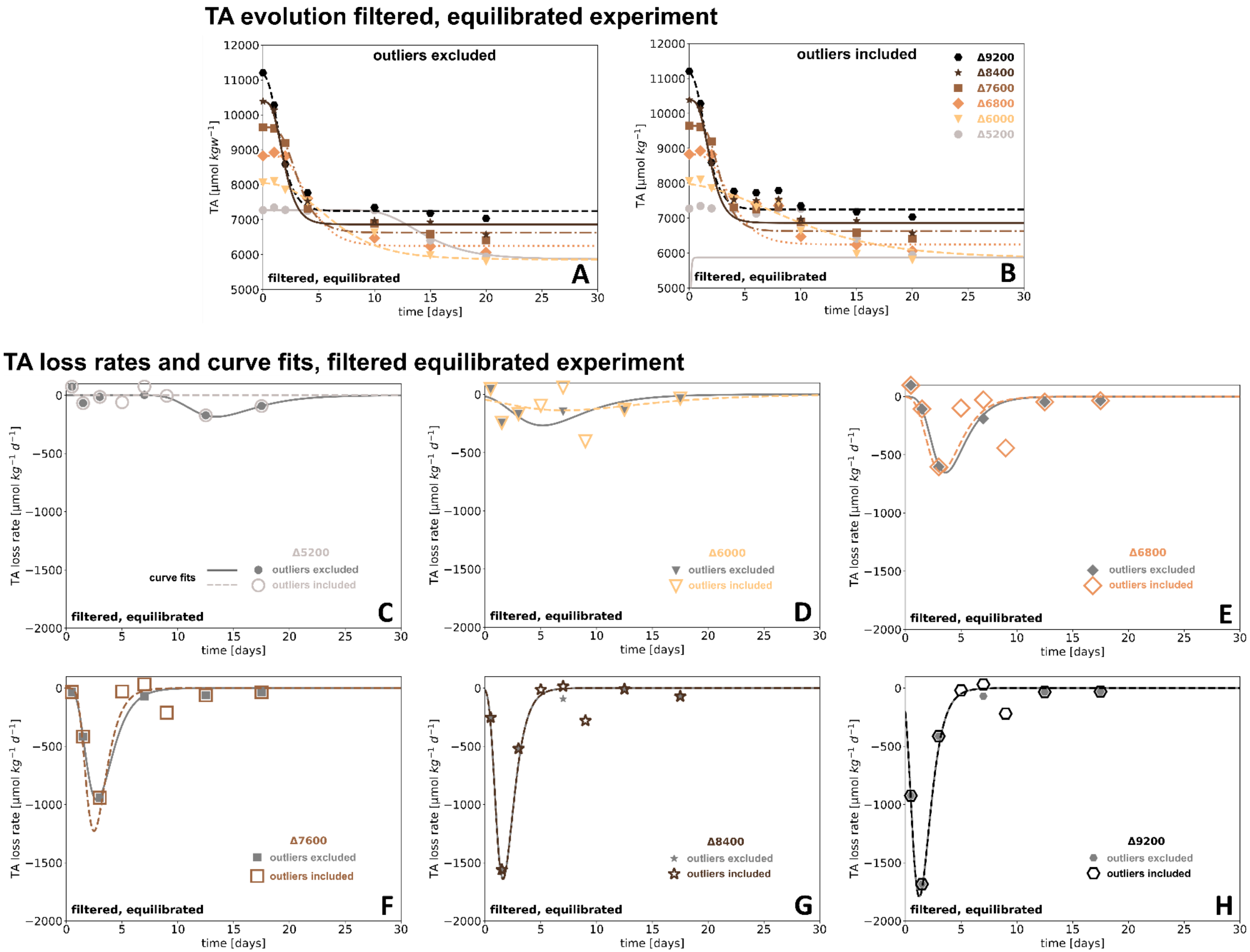

Figure R1: Comparison of TA evolution, TA loss rates, and corresponding curve fits with and without outliers, in the filtered equilibrated treatments. TA evolution excluding outliers (A) and including outliers (B). (C-H) TA loss rates and curve fits for each treatment level entering an accelerated precipitation phase, solid gray markers and lines: without outliers, hollow markers and dashed lines: outliers included. Curve fits differ in case outliers appear to fall into the accelerate precipitation phase of a treatment level, excluding the outliers was met by averaging the rate between the last and first measurement point before and after the period with outliers (days 6 and 8)

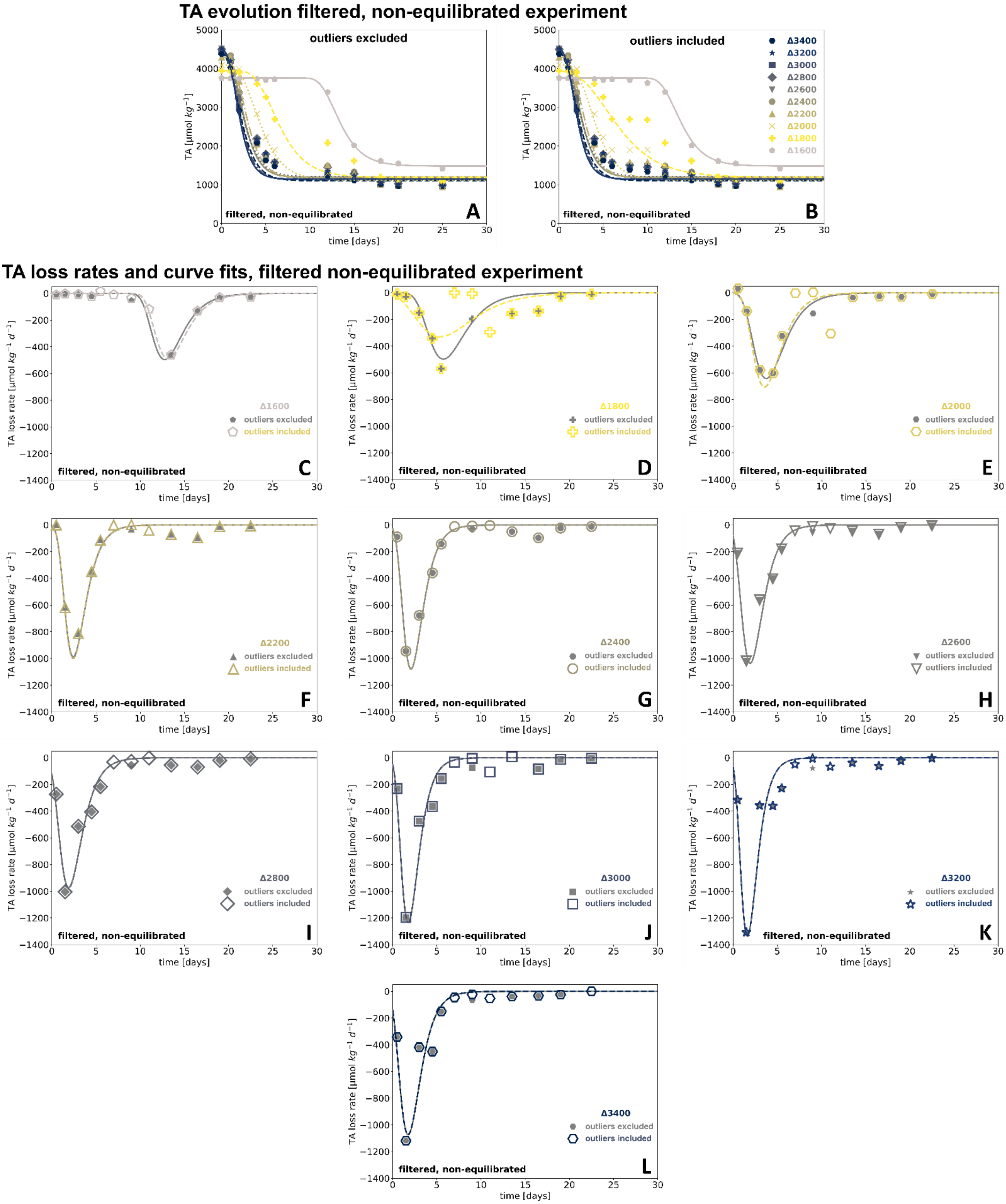

Figure R2: Comparison of TA evolution, TA loss rates, and corresponding curve fits with and without outliers, in the filtered non-equilibrated treatments. TA evolution excluding outliers (A) and including outliers (B). (C-L) TA loss rates and curve fits for each treatment level entering an accelerated precipitation phase, solid markers and lines: without outliers, hollow markers and dashed lines: outliers included.

Comparison of carbonate precipitation kinetics—with and without outliers

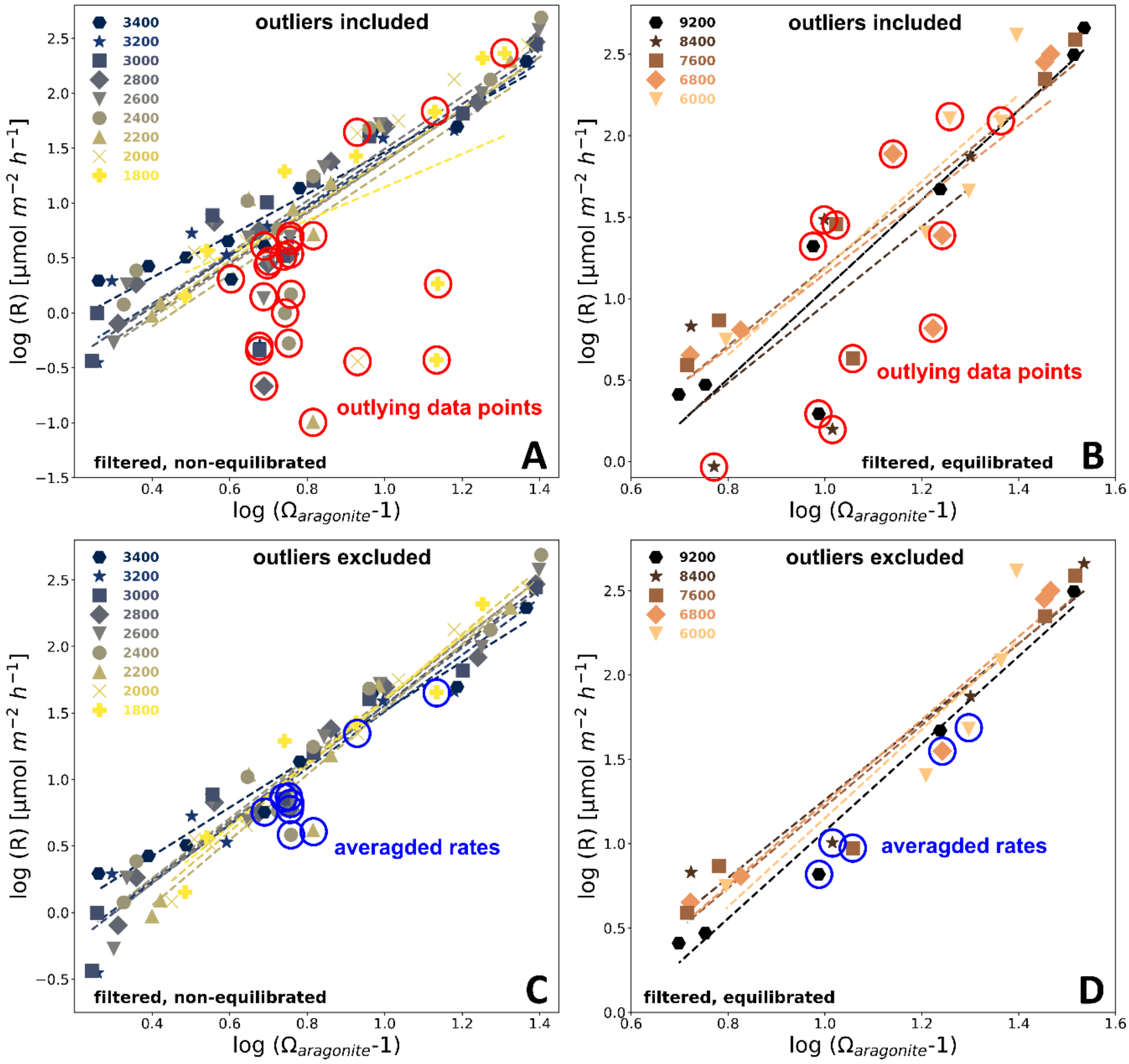

Figure R3: Comparison of carbonate precipitation kinetics for filtered treatments that entered the APP; (A) filtered neq with outliers, (B) filtered eq with outliers, (C) filtered neq without outliers, (D) filtered eq without outliers, red outlined data points: excluded outliers, note that some outliers exhibited an increase in TA during the precipitation process and could therefore not be plotted on a logarithmic scale; blue outlined data points: rates representing the average rate calculated between the last and first datapoint before and after the precipitation halted.