

Report #1

Submitted on 11 May 2021

Referee #1: Jens Daniel Müller, jensdaniel.mueller@usys.ethz.ch

Anonymous during peer-review: Yes **No**

Anonymous in acknowledgements of published article: Yes **No**

Recommendation to the editor

1) Scientific significance

Does the manuscript represent a substantial contribution to scientific progress within the scope of this journal (substantial new concepts, ideas, methods, or data)?

Excellent **Good** Fair Poor

2) Scientific quality

Are the scientific approach and applied methods valid? Are the results discussed in an appropriate and balanced way (consideration of related work, including appropriate references)?

Excellent **Good** Fair Poor

3) Presentation quality

Are the scientific results and conclusions presented in a clear, concise, and well structured way (number and quality of figures/tables, appropriate use of English language)?

Excellent **Good** Fair Poor

For final publication, the manuscript should be

accepted as is

accepted subject to technical corrections

accepted subject to **minor revisions**

reconsidered after **major revisions**

I am willing to review the revised paper.

I am **not** willing to review the revised paper.

rejected

Suggestions for revision or reasons for rejection (will be published if the paper is accepted for final publication)

The authors did a good job in implementing the comments made by both reviewers, with one substantial exception being the revision of the confidence interval of the observed pH changes over time.

[We thank Dr. Müller for the careful re-review of the manuscript and we address the comments and suggestions below.](#)

In the first submitted version of the manuscript, the authors stated that:

"A linear regression on all pH measurements [...] has a slope of $-0.0058 \pm 0.0008 \text{ yr}^{-1}$ (mean \pm 95% C.I.)."

In the revised version of the manuscript, the CI of the slope was changed and additional information is given:

"A linear regression on all pH measurements [...] has a slope of $-0.0058 \pm 0.0011 \text{ yr}^{-1}$ (mean \pm 95% C.I.). The upper and lower bounds of ΔpH at $t = 365$ days, -0.0042 and -0.0076 , are important to consider when utilizing this bagged storage method of tris. These bounds provide the broadest expected range in pH change over a year of storage, and include both the intercept and slope confidence intervals."

There are several follow-up questions and recommendations to this change, which I suggest the author take into account:

- Why did the numeric value of the CI increase, while the grey shaded area in Fig. 3 is now narrower than in the first version?
- Which version (first submission or resubmission) of the numeric CI value in the text and the CI interval in Fig. 3 are correct?
- The fact that the grey shaded area in Fig. 3 is now narrower appears contradictory to the statement that it represents the "broadest expected range in pH change".

The confidence interval in the first submission was calculated from the mean of replicate measurements. The confidence interval in the resubmission was calculated from all individual measurements, resulting in a slightly larger confidence interval.

The resubmission has the correct value of the numeric CI in the text and CI in Fig. 3.

In the original submission, we misused the confidence interval function in MATLAB resulting in an incorrect exaggeration of the illustrated wedge and a mismatch between the reported numeric range and the illustrated range in the wedge in Fig. 3. We corrected this mistake which results in the new values as reported and illustrated in Fig. 3 in the resubmission. This correction now provides the broadest expected range in pH.

- Please clarify how the CI of the slope and the intercept were combined to derive the upper and lower bounds.

Both the slope CI and intercept CI contribute to the upper and lower bounds.

The following text is added to clarify:

These bounds provide the broadest expected range in pH change over a year of storage and include both the intercept and slope confidence intervals (*slope_{CI}* and *intercept_{CI}*, respectively). For example, the upper bound of ΔpH at $t = 365$ days is calculated as:
upper bound = (*slope* + *slope_{CI}*) * 365 + *intercept* + *intercept_{CI}*.

- Finally, I repeat my suggestion to include the information contained in the upper and lower

bound estimates also in the abstract. It is likely among the most relevant piece of information for many readers.

Added the following text to the abstract:

The upper and lower bounds of expected pH change at $t = 365$ days, calculated using the averages and confidence intervals of slope and intercept of measured pH change vs. time data, were -0.0042 and -0.0076 from initial pH.

Additional technical comments are:

L.136ff: “To account for pH-dependent errors from impurities in unpurified mCP, a pH-dependent correction factor was determined based on the protocol outlined in Takeshita et al. (in review). Briefly, pH of seawater was measured subsequently using impure dye (pHimpure; from Aldrich, lot MKBH6858V) and purified dye (pHpure; from Robert Byrne’s Lab, University of South Florida (Liu et al., 2011)) over a range of pH between 7.4 to 8.2 at approximately 0.2 intervals. Varying ratios of tris:trisH⁺ were used to obtain different solution pH ...”

With the new information included in the revision it is now clear that tris buffered solutions were used to determine the impact of dye impurities, and not seawater solutions. Thus, I agree with the applied correction procedure. However, please replace:

- “pH of seawater was measured” with “pH of tris buffered artificial seawater solutions was measured”.

Replaced with “Briefly, pH of natural seawater with different ratios of added tris:trisH⁺ was measured...”

- “review” with “review”.

Replaced.

- “Lab” with “laboratory”

Replaced.

In Figures 2 and A2, I suggest to avoid redundant information to make the figures more easy to read.

Fig 2:

- Remove labels in plot, as they are redundant with the color/shape.

We disagree with the reviewer on this point, believing that they make the plot easier and quicker to understand. We have changed the label and slope order to make it easier to read.

- Remove “- shown by marker XXX” legend

Removed.

- In the caption, consider changing “Bag type 1 is shown in blue (light blue for the 185damaged bag of type 1), 2 in orange and bottle in green. Tris batch 1 is depicted as circles, 2 as squares, 3 as diamonds and 1864 as stars. Storage location in tank has a black fill and lab symbols have no fill.” to “Storage vessel is indicated by color, storage location by fill, and Tris batch by color”

Replaced with “Tris batch is indicated by shape, storage vessel by color, and storage location by fill.”

Fig A2:

- Shapes and fill in the legend are not readable. Consider using the same legend as in Fig. 2.
Replaced with Fig. 2 legend.

Thanks again to all authors for developing and testing this new approach to perform quality control of pH measurements under field conditions. I hope your findings will be adapted rapidly.

Thank you for all the feedback on the manuscript.