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Title: Metrological concepts applied to Total Alkalinity measurements in seawater: reference materials, inter-laboratory comparison and uncertainty budget

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Response to the Editor

Dear Editor Mario Hoppema,

We would like to greatly thank you for taking the time to review our manuscript, and giving us the opportunity to improve it thanks to your valuable comments and suggestions. We have carefully considered all your editorial comments; they have been integrated in the reviewed manuscript.

Response to reviewer – RC1

Dear reviewer,

We would like to greatly thank you for taking the time to review our manuscript this second time, and giving us the opportunity to improve it thanks to your valuable comments and suggestions. We have carefully considered all your comments. You will find below how we addressed them in the revised manuscript.

The line numbers correspond to the reviewed manuscript with changes marked.

Line	Comment	Response
General comment	The reviewer recommended using an LLM-based AI to improve the language. Technically, the topic is presented properly now. I leave it to the editors to assess language.	This comment is considered addressed.
21	Remove "potentially". Either a result is traceable to a metrological reference or it isn't. There is no in-between status.	The manuscript has been changed accordingly.
56-57, 731	The reviewer is not convinced that simply having no uncertainty budget is the sole reason why TA measurements using the conventional method are not traceable (to whatever reference). The method relies on several measured quantities the traceability of which may not be fully established or may even be inconsistent. For example, the total hydrogen ion concentration is quantified through pH/potential measurements using glass electrodes. However, what is the metrological refence of those results? Primary pH buffers, the values of which include or do not include the Bates-	The introduction has been changed as follows: "However, the RMs distributed aren't fully traceable **partly** due to the fact that they aren't given with a rigorously assessed uncertainty. **Other traceability issues coming from the measurement process should also be carefully investigated**". This has also been specified in the traceability section.

	Guggenheim convention? In fact, the pH of those buffers is defined in terms of activity, while Dickson's guide assumes the potentials are a measure of H+ ion concentration. Moreover, how are liquid junction potentials of the glass electrode considered, which also affect the measured potentials significantly? Those are difficult questions to be answered in assessing traceability of the TA measurement procedure. That said, the reviewer does not intend to question the overall paper on the basis of these traceability concerns. However, it would be expected that this point is acknowledged in the introduction and the traceability section as an open issue. In fact, it even supports the value of the proposed artificial RM.	
172	Correct: The measurement result~~s~~ at zero NaCl mol kg- 1 sol is shown in Fig. 1 and support**s** this reasonable assumption. Or: The measurement results at zero NaCl mol kg- 1 sol ~~is~~ are shown in Fig. 1 and support this reasonable assumption.	The manuscript has been corrected accordingly.
176	"The measurements presented in Fig. 1, which can question the linear behaviour… " I would suggest to write "Linearity of the measurement results is a rough assumption that is further discussed in section 3.3.2."	The manuscript has been changed accordingly.
287	"This study highlighted that the determination of the homogeneity is highly dependent on the variability of the measurement method." Should rather be "This study highlighted that the robustness of the determination of the homogeneity is highly dependent on the variability of the measurement method.	The manuscript has been changed accordingly.
289, 464	Add reason: "Uncertainty resulting from within-bottle inhomogeneity can usually be neglected for liquid refence materials." A quantitative number should be added to the discussion in 3.4.4 to support the statement that ", the (1/3) criterion was slightly exceeded", i.e. how do the measurement repeatabilities of the three batches, which can be expressed by M-within, compare to the target uncertainty.	The reason given has been added. The M_within value has also been added in the discussion section, being of 1.5µmol/kg, while the targeted uncertainty is 2µmol/kg.

436	Again, remove “potential”. The issue of the TA- background has been appropriately discussed. It is indeed an issue that must be addressed. But it is not so significant that SI traceability of the assigned TA value must be stated as “potential”. It is rather an uncertainty of the uncertainty.	The manuscript has been changed accordingly.
460	Replace “precision” by “repeatability”.	The manuscript has been changed accordingly.
Section 6	I see the necessity, but I am not yet convinced that the proposed concept of a kind of “practical traceability” using two RMs having different traceabilities has been developed in sufficient metrological depth. However, the issue is not fundamental in the context of the paper (even though it is very fundamental in general). Thus, I consider it resolved.	This comment is considered addressed.
486	? Why? This comment didn’t request a change. I was just appreciating the observation.	“was less stable” was changed to “lack stability” in the previous round of revision. This didn’t change the meaning of the sentence.

Response to reviewer – RC2

Dear reviewer,

We would like to greatly thank you for taking the time to review our manuscript this second time, and giving us the opportunity to improve it thanks to your valuable comments and suggestions. We have carefully considered all your comments. You will find below how we addressed them in the revised manuscript.

The line numbers correspond to the reviewed manuscript with changes marked.

Line	**Comment**	**Response**
166-167	Line 165: Instead of stating "using the materials and devices presented in Appendix B" and leaving the reader to search for this information in a supplementary section and losing their train of thought, it would be clearer to state, for example, "using the HCl standardized at SMU and a the Metrohm titration system as described in Appendix B."	The manuscript has been changed accordingly.
815	Line 802: Metrohm is misspelled.	The manuscript has been changed accordingly.

577	Line 567: "The precision of the method, sR..." The authors did not address my previous comment here. They introduce a new notation sR that is not used in the other equations. (Equations 15 and 16 use sr for intra-laboratory standard deviation, which is different from sR.) The authors define sR as "precision...given by computation of sL and sr," but they don't describe exactly how sR is computed. Based on Table 5, it appears that sR is computed by summing sL and sr in quadrature (i.e., sR = sqrt(sL^2+sr^2) = 1.99 umol/kg as the authors report in the text).	The notation "sR" has been removed to avoid confusion.
Throughout the manuscript	Scientific notation: In various places in the manuscript, the authors have still left numeric values in E+XX notation rather than in scientific notation.	All values has been changed for scientific notation.
340-347	Fig 2 is a new addition in the revised manuscript. I would recommend including the results of Batch 1 on this graph so the stability of the two batches can be compared.	Figure 2 has been updated accordingly.
Throughout the manuscript	Expanded uncertainties and coverage factors: Although it is acceptable to report expanded uncertainties with a coverage factor of 2, it may not always represent a 95% level of confidence, depending on the effective degrees of freedom which was why I recommended the authors report this information. It can be included in the supplementary section. It may also be acceptable to state that the coverage factor of 2 corresponds approximately to a 95% level of confidence if this assumption is true for the authors' estimates.	It has been specified that the coverage factor of 2 corresponds approximately to a 95% level of confidence.