

**June 06 2024 Editor comments:**

**Public justification (visible to the public if the article is accepted and published):**

*Dear Editor,*

*Thank you very much for your time, work, and the helpful comments and suggestions, which really improved our manuscript. We significantly edited the manuscript in accordance to your suggestions. We reworked the Introduction and reduced the Discussion in length. We also rephrased many sentences to increase the readability.*

Dear authors,

There is obviously a big difference between the two subregions around Ameland and the Ems estuary. This should be made more clearly and separated in the manuscript.

*AC: We have addressed this concern and have reworked several parts of the manuscript to make this point more clear.*

The Introduction still needs work and restructuring, even if some changes were already made after previous rounds of reviews.

*AC: We have reworked the Introduction.*

It is mentioned that two (half) tidal cycles were sampled and measured. However, data of only one of these are shown and discussed. Do the other data show the same features or are they quite different? This is important info which must be provided. Please mention somewhere what the other data look like, how they compare with the presented data and whether the conclusions are supported by the second half tidal cycle.

*AC: We added a statement in the Methods section saying that we only used the second half tidal cycle (flood tide) for a data comparison and to relate whether the ebb tide data are in a similar range. We added the flood tide data in the Appendix.*

A previous review objected against the use of contents instead of concentration. However, the use "contents" instead of concentration is allowed. Maybe cite the paper by Jiang et al (2022, Front Mar Sci), to make this clear.

*AC: We have changed it back to the use of concentration that we already used in the first version.*

The notation 2400  $\mu\text{mol kg}^{-1}$  TA, i.e., with TA at the end is not common and may be confusing. Please change it throughout the manuscript for all concentrations/contents.

*AC: We had this notation in the first version of the manuscript and changed it back to  $\mu\text{mol TA kg}^{-1}$  etc.*

List of minor and technical issues:

L10 "The oceanic buffering capacity total alkalinity (TA), as the major global CO<sub>2</sub> sink, is of growing scientific interest". This is a strange contention and not necessarily true. Please change, for example, by just saying that TA is an important chemical property which plays a role in oceanic buffering capacity.

*AC: We agree, and changed the sentence according to your suggestion.*

L11 "... generated by chemical weathering on land ..." Add: on land.

*AC: Done.*

L12-13 This study shows observations of TA, ... instead of "This study observed TA, ..."

*AC: Done.*

L18-19 "We assume that seasonality and the associated nitrate availability in particular influence TA

generation by denitrification, which we assume is low in spring and summer.” This is too much of assumptions in an abstract. Please only present the facts.

*AC: We have rephrased this part.*

L24 “The (still unofficial) Anthropocene describes ...” Or something similar, as the Anthropocene has not been acknowledged officially.

*AC: We added a ‘so called’ and “Anthropocene”.*

L25-26 “The climate and the increasing atmospheric CO<sub>2</sub> content is mainly regulated by the open ocean.” This is not correct. The biosphere takes up more anthropogenic CO<sub>2</sub> than the oceans – see the Global Carbon Budget.

*AC: We removed the ‘mainly’.*

L26 The around 30% is more like 25% (see Global Carbon Budget).

*AC: We have corrected it.*

L32 susceptible to what? Please be more precise.

*AC: We added “to changes”.*

L49 strong tidal currents instead of: high tidal currents

*AC: Done.*

L55-57 “Understanding of TA sources have recently become increasingly important due to increasing anthropogenic CO<sub>2</sub> emissions, and the resulting demand for ocean based net-negative CO<sub>2</sub> emissions” This needs a short explanation as the connection between the two is unclear.

*AC: We rearranged this sentence into two and added a connection to make it more comprehensible.*

L74 Please use format like 21 May 2019

*AC: Done.*

L76 delete: continuously

*AC: Done.*

L80 Add solution after chloride

*AC: Done.*

Section 2: When were the samples measured, i.e., how long after sampling? Please add this to the different methods.

*AC: We have added the reference material and the month when the samples were measured.*

L97 I guess you mean: ... both with a measurement precision ...

*AC: Yes, thank you. We adjusted the sentence.*

L97 It is kind of strange that you cite the precision with a different study. Did not you yourself determine the precision and accuracy?

*AC: This mentioned study references the methods and the official given precision of the instrument.*

L98-99 “To ensure a consistent calibration of both measurements, certified reference material (CRM batch # 187) provided by Andrew G. Dickson (Scripps Institution of Oceanography) was used” How was this CRM used? Did you adjust the measured data?

*AC: We added the missing information. CRM were measured before and after the samples and used for data drift correction.*

Section 2.3 What is the precision and accuracy of the nutrient measurements? Were any CRMs used?

*AC: We have added the used reference materials and the max. standard deviation.*

L117 change to: (in particular Hoppema, 1990)

*AC: Done.*

L117-118 “ ... we observed TA and related parameters from the coastal mainland towards the open North Sea as a surface water transect” (delete: “the spatial distribution of” because that was already mentioned earlier in the sentence.

*AC: Done.*

L119-120 „Salinity was relatively stable with only minor differences varying from 28 to 33“ First, I think stable is the wrong term here. Second, this large range does not indicate a homogeneous salinity but rather significant variations. The range of salinity is obviously larger than this, as values in the Ems are smaller. Please rephrase the whole paragraph.

*AC: Done.*

L123 “oceanic” is not the correct term here. The North Sea is not an ocean. Actually, “In contrast to the oceanic side,” may as well be deleted as it has no function here.

*AC: We changed all the “oceanic” into North Sea side.*

L124 “Only in the Ems Estuary, the contents were even higher” Delete: Only. Also close to Harlingen TA is higher.

*AC: Done.*

L126 support

L126 “supporting the assumption of TA being generated in this tidal flat area.” It is too early for this contention. It may as well be the discharge of river water that causes high TA.

*AC: We removed this sentence.*

L127 “showed a similar pattern” Similar to what? Please phrase more precise.

*AC: We removed this part.*

L127 Use other word for “ocean” Please also change this at other places in the text.

*AC: Yes, we did this and used just the term “North Sea”.*

L129 I think the silicate contents are given with one digit too much, as the accuracy is probably not that high (accuracy has to be stated in the Methods section)

*AC: Done.*

L130 not site, rather region

*AC: Done.*

L134 delete: similarly

*AC: Done.*

L135 region instead of transect. There are a few transects shown in this study.

*AC: Done.*

L144 “The strong impact from the inner Ems Estuary is visible in all parameters with higher values in the outer estuary and its adjacent zones” This is an awkward sentence. Please rephrase.

*AC: Yes, we rephrased this paragraph.*

L145 higher values of what?

*AC: We rearranged this whole paragraph.*

L148 in the mixing plot between TA and salinity. Add: plot, namely TA and salinity do not mix themselves.

*AC: Done.*

L148 “A relatively linear mixing behavior” Relatively is awkward here. Please rephrase

*AC: Done.*

L150 “identifying the Dutch Wadden Sea as a source of TA” This is not correct. A linear relation in an estuary in this case shows that the TA of the river water is high.

*AC: This is correct. This part of the sentence was only related to the Vlie Inlet. However, we rearranged the paragraph since it led to misunderstanding.*

L150-151 “In contrast to the TA content computed for the salinity end-member in the Ems-Dollard Inlet, we detected higher TA contents around Ameland” This sentence is strange; please be more precise.

*AC: Both this sentence and the whole paragraph were rearranged.*

L154 The values are not increasing. There is just a range of values at constant salinity.

*AC: Done.*

L156 (figure caption) Change to: Mixing plot of total alkalinity (TA) and salinity ...

*AC: Done.*

Figure 3 Please indicate whether the correlations shown are statistically significant or not.

*AC: Done.*

L169 "DIC contents were similar to TA" This is of course not correct, as the data show. This is about the development/course of DIC contents.

*AC: Yes, we adjusted this sentence to make it clearer.*

L172 "Nitrate concentrations approached seawater concentrations" What are seawater concentrations for nitrate? These are highly variable. Please be more precise in what you intend to say.

*AC: We have reworked this part.*

L173 delete: slightly

*AC: Done.*

L183 Change to: the maximum pH was 8.07 at high tide

*AC: Done.*

L188 (caption) This is only half a tidal cycle. Maybe it is good to mention the local time difference with UTC

*AC: Done.*

Figure 4: Two important variables are missing, namely salinity and temperature. These must be added here for better understanding of the tidal cycle.

*AC: Done.*

L193 "The 192 strong tidal forcing induces a strong benthic-pelagic coupling" This needs a reference.

*AC: Done.*

L194 hypothesis instead of assumption appears to be the better choice here.

*AC: Done.*

L201-202 "Based on the correlation of TA and silicate ( $R^2 = 0.93$ ), and on the nonlinear relation between both, TA and salinity ( $R^2 = 0.32$ ), as well as silicate and salinity ( $R^2 = 0.21$ )," How can a non-linear relation have a correlation? And is this correlation significant or not? This must be rephrased to something like: Based on the correlation of TA and silicate ( $R^2 = 0.93$ ), and on the insignificant relation between TA and salinity ( $R^2 = 0.32$ ), as well between as silicate and salinity ... .

*AC: This is why we named it non linear relation (and not correlation), but we agree that this can be misleading and adjusted the sentence accordingly to your suggestion.*

L203 "Both TA and silicate increased almost proportionally" What does this mean? Please describe more clearly.

*AC: We have rearranged this sentence.*

L204 Change to: non-conservative behavior of TA with respect to salinity and silicate to salinity ...

*AC: Done.*

L207 "from high tide to low tide" I guess you just mean the half tidal cycle as treated above. As now, this is confusing.

*AC: That is correct, so we added "the half tidal cycle".*

L208 "recommend" here is awkward. And why would you ignore these data? This sounds quite arbitrary. Please give a good reason for that.

*AC: These data are not ignored in the analyses. We just wanted to highlight that the first points are on the tipping point between the change in tide. We deleted the confusing part of the sentence and reworked this section.*

L209-210 "First, the correlation between TA and DIC reveals the ratio between anaerobic and aerobic

processes, which identifies a strong positive correlation between DIC and TA ( $R^2 = 0.93$ ) with TA contents higher than DIC contents (Fig. 5a).” This is not well phrased. Change to something like: The correlation between TA and DIC is a measure for the ratio between anaerobic and aerobic processes. Our data show a strong positive correlation between DIC and TA ( $R^2 = 0.93$ ) with TA contents higher than DIC contents (Fig. 5a).

*AC: Thank you for this suggestion. We arranged the sentence accordingly.*

L210-212 “However, even with contents of TA higher than DIC, the slope of 1.87 indicated DIC release excess with an increase in DIC ( $\Delta\text{DIC} = 101.3 \mu\text{mol kg}^{-1}$ ) almost twice as high as TA ( $\Delta\text{TA} = 51.6 \mu\text{mol kg}^{-1}$ ) (Fig. 5a).” This sentence is unclear. Please rephrase so that it becomes clear what is meant here.

*AC: Done.*

L214 delete: almost

*AC: Done.*

L218-219 “suggest a stronger effect of TA generation than nitrate production”. The word “effect” can be omitted here as it is confusing: ... suggest a stronger TA generation than nitrate production ...

*AC: Done.*

L239 insert possible or probable before TA

*AC: Done.*

L243 add something like: ...similar range, but the spatial gradients are opposite.

*AC: Done.*

L244-246 “The conservative mixing in the Vlie Inlet can be explained by the fact that more North Sea water pass through the deeper inlets and transport more seawater towards the coast.” This is not clear to me. How does this explanation work?

*AC: We have rearranged this sentence.*

L257 The salinities observed were not constant. They varied possibly not that much, but that is not the same as constant. Please rephrase.

*AC: Done.*

L264 hypothesized instead of: assumed? Did they find indications for it?

*AC: We have changed it into “indicated”.*

L266 “from the early 1990s” The study was actually from the late 1980s.

*AC: Done.*

L268-269 “Our present TA data and the historical TA data show no large differences in the range of values observed during a tidal cycle.” This sentence is strange. What about: Comparing our present TA data and the historical TA data, there is not a large difference in the range of values observed during a tidal cycle.

*AC: Done.*

L269-270 “However, an in-depth interpretation and comparison of both data sets would exceed the capacity of these data, leading us to focus on TA generation during our cruise” I do not understand this sentence. What is the capacity of these data?

*AC: Here, we mean the limited number of TA data. We have rephrased this sentence.*

L275 estimated instead of assumed. It would be good to add that they used a model for this at this place.

*AC: Done.*

L276-277 “However, an inclusion of our TA export into the model used by Schwichtenberg et al. (2020) would be unreliable, since our TA export based only on one tidal observation” Why should that be unreliable? It can be just part of that estimate, as the present data are observed data. However, if you would use the present tidal data for estimating the entire TA export, that indeed would be unreliable.

*AC: Yes, that is what we wanted to say. We have rearranged these sentences.*

L283 “In order to gain further insight into potential sources of TA, we included nutrients in our investigation” This sentence is too general. Earlier in the manuscript, nutrients were already shown.

*AC: We have rephrased this sentence.*

L285 Write: Van der Zee, or van der Zee

*AC: Done.*

L283-285 “The main focus was on silicate that we used as a natural tracer since it is not directly provided anthropogenically and allowed us to determine the silicate source” This sentence is unclear; please be more precise what you want to convey. The authors base this contention on a reference from 2005. Since the early 2000s the situation in the Wadden Sea could have changed a lot, as was also found for the sources of alkalinity. Do you have any indication that nothing has changed?

*AC: We have rewritten this sentence to clarify why we used dissolved Si as a tracer for pore water exchange and substantiate this with early work on Si dynamics in the Dutch Wadden Sea. Details on pore water data were later published by Rutgers van der Loeff (1980) but already used by van Bennekom et al. (1974). The question whether Si dynamics have changed is valid. River data do not show a large change in dynamics, but in our opinion, an in-depth discussion of the Si long-term dynamics is beyond the scope of this article.*

L288 hypothesis instead of assumption

*AC: Done.*

L290 “salinity consistently being above 32” It is not clear which area is concerned here. The Ems estuary has lower salinities, but also the Vlie has a salinity of about 30 (Fig.3). Please be more precise.

*AC: We have added the sampling area (around Ameland island).*

L291-292 “This can be supported by the relation between silicate and salinity in which we observed a 291 non-conservative behavior (Table B1). Since TA behaves also non-conservative relative to salinity (Table B1)” This says actually the same as the previous sentence. In addition, Table B1 does not show a relation at all; it just shows data. You may just refer to Table B1 after the previous sentence.

*AC: We have rearranged both sentences.*

L297-298 delete: “In May, they observed low salinities indicating freshwater. However, in September they observed constant marine salinities referring them to be exclusively composed of recirculating seawater.”

*AC: Done.*

L298-299 “Since we constantly observed marine salinities,” This is an awkward sentence. Change to something like: Since we observed relatively constant salinities ...

*AC: Done.*

L307 “smaller than” instead of: below

*AC: Done.*

L308 delete (twice) clearly

*AC: Done.*

L311-313 One can write this much easier and understandable. Please rephrase.

*AC: Done.*

L313-314 “However, when silicate occurred dissolved in water it does not contribute to TA generation (Meister et al., 2022).” I do not understand this. Please explain .

*AC: This sentence was deleted while reworking this part.*

L314-315 “A TA excess related to silicate was also observed in the correlation between TA and silicate (Fig. 5b).” Of course, you can see this in Fig. 5b. These are the same data.

*AC: We have deleted this sentence and added the ref of Fig 5b to the first sentence.*

L318 Use the Greek symbol for omega. Actually, “With the observed omega values,” can be deleted because the same is written later in the sentence.

*AC: Done.*

L321 “because of the  $\Omega$  supersaturation of the overlying water” This is not the reason for not being able to determine TA generation in the sediments, so please delete.

*AC: Done.*

L326-327 This is an anacoluthon. Please correct the sentence. Which  $\Delta$ TA and  $\Delta$ DIC is this? They have not been defined.

*AC: Done, we have rephrased this paragraph.*

L328 increase of DIC. Which increase? Again,  $\Delta$ DIC was not defined. Please be more precise.

*AC: We added “during ebb tide” to show that this is the increase of DIC during ebb tide.*

L330 “high CO<sub>2</sub> export” Which export, from where to where?

*AC: Since the word export was misleading, we have changed it into production.*

L330 delete: also, as this is not observed in this study. It is only hypothesized.

*AC: Done.*

L335 continues instead of: continuous

*AC: Done.*

L3345 exceeds instead of: exceeded

*AC: Done.*

L353 less instead of: lower

*AC: Done.*

L357 “due to high turnover rates” I would say: due to high photosynthetic activity

*AC: Done.*

L373 It would be a nice service to the reader and make the paper more readable if the chemical reaction scheme of aerobic OM respiration with the associated formation of ammonium would be shown here.

*AC: In order to reduce the discussion in length as suggested, we removed this whole part of the discussion and reworked it.*

L375 delete: leading to ammonium formation. This was already written at the beginning of the sentence.

*AC: Done.*

L376 increases instead of: would increase

*AC: Done.*

L388-389 “the simultaneous increase of TA and nitrate is noticeable for us, because nitrification consumes TA” It is unclear what the authors try to convey here. Please rephrase and explain.

*AC: We have rearranged this sentence.*

L391 “Low nitrate concentrations resulting in a reduced availability of bound oxygen, i.e., electron acceptors.” This is not a correct sentence. Please rephrase.

*AC: We removed this sentence and reworked this whole paragraph to reduce it in length, as suggested.*

L401 define POC, as it is used for the first time

*AC: Done.*

L413-414 “While observing the spatial TA distribution and TA generation” This manuscript does not observe TA generation per se. It only deduces it and speculates about it.

*AC: We have added “deducing” to this sentence.*

L415 delete: clearly

*AC: Done.*

L420 deduced instead of: identified

*AC: Done.*

L420 and further: It should be made clear that this was not observed but rather deduced with some speculation.

*AC: Done.*

L428 Spatial instead of: Latitudinal and longitudinal

*AC: Done.*

L431 Table B1 It is half a tidal cycle. Please mention the position in the table header. An obviously missing variable in this table is: time.

*AC: We added both.*

L432 Format 21 May 2019

*AC: Done.*

L437 Which other data were measured during the cruise? At least it was mentioned that there is a second (half)tidal cycle. The data should be published along with the manuscript. Other data, which is probably not that much, could be published as supplementary material.

*AC: During this cruise we only measured the analyzed data and the other half tidal cycle, which we used to compare and relate the range of data from the ebb tide tidal cycle. We added these data in the Appendix as Table B2.*

Additional private note (visible to authors and reviewers only):

This manuscript has had a few rounds of reviews already, but it still needs work to get it into shape for final publication. I have listed below quite a long list of mostly technical issues, and the list is not even complete. The writing is partly sloppy and should be much improved. I encourage the senior authors to thoroughly go through the manuscript again. At many places the phrasing is not precise enough for a scientific study. Sometimes the reasoning is hard to follow. In many places the text is overly drawn out and it can be more succinct.

As to the science, I think the data and its discussion are worthy of publication. However, the discussion on the alkalinity sources is in many aspects speculative and as being speculative, it should be reduced in length. Note that there is not a single measurement that could confirm the outcome of the analysis. Thus, the authors should tone down the firm conclusions, even if there are indirect indications that the tidal flats may be important sources of TA. I would still want to give the authors the chance to improve the manuscript and make the major revisions that I am here suggesting. Please note that all comments must be addressed satisfactorily and if the authors decide not to apply the changes, they need a good reason for that.