Carter et al. introduce a new composite tracer Alk* in order to `isolate the impact of acidification on biological calcification and remineralization' (abstract). Total alkalinity (A_T) in the ocean changes due to several processes: (1) evaporation/precipitation, (2) formation of organic matter by phytoplankton, (3) remineralization of organic matter, (4) formation/dissolution of $CaCO_3$, (5) redox processes in marine sediments and in the water column, (6) riverine input of water with varying alkalinity (usually, however, not always smaller than mean seawater concentrations). Carter et al. are especially interested in (4) formation/dissolution of $CaCO_3$, which is difficult to quantify for the world oceans. They propose a new tracer (Alk*) that should ideally be a tracer of $CaCO_3$ cycling alone.

The definition of Alk* is a bit more tricky than for other *-quantities:

Alk* = A_T + 1.26 * [NO_3^-] – S * mean(A_T + 1.26 * [NO_3^-]) / mean(S)   {Alk*def}

where the mean values are calculated over the `top 50 meters of the ocean´ (actually averaged vertically as well as horizontally yielding a value of 66.4 μmol kg^{-1} for mean(A_T + 1.26 * [NO_3^-]) / mean(S), compare Eq. 5), A_T is the total (or titration) alkalinity and S is the salinity. The term 1.26 times the nitrate concentration takes care of A_T changes due to formation of organic matter by phytoplankton (uptake of nitrate, phosphate, sulphate) or the remineralization of organic matter. The value of the coefficient can be either taken from observations (1.26 based on Kanamori and Ikegami, 1982) or derived from the stoichiometry (N:P:S; Redfield & extensions; for details compare Wolf-Gladrow, D. A., Zeebe, R. E., Klaas, C., Körtzinger, A., and Dickson, A. G.: Total alkalinity: the explicit conservative expression and its application to biogeochemical processes, Marine Chem., 106, 287–300, 2007.) of phytoplankton.

The sum of the first two terms on the rhs of {Alk*def} is called 'potential alkalinity' A_p:

A_p = A_T + 1.26 * [NO_3^-]

A_p is a conservative quantity with respect to organic matter production based on nitrate uptake (decrease of nitrate & increase of A_T) and remineralization of organic matter (inclusively oxidation of reactive nitrogen to nitrate).
Please note that Alk* can take on positive as well as negative values. The surface ocean mean (upper 50 m) of Alk* is zero by definition. The depth of 50 m used for the definition of Alk* is rather arbitrary and in my opinion a weakness of the proposed Alk* concept.

The discussion on regional variations of A_T and Alk* and the influence of riverine alkalinity input is quite insightful. Another interesting region with 'unusual' alkalinity values is the Mediterranean Sea (compare, for example, Schneider, Anke, Douglas WR Wallace, and Arne Körtzinger. "Alkalinity of the Mediterranean Sea." Geophysical Research Letters 34.15, 2007).
I found it difficult to grasp the content of the section on process importance on saturation level Ω (I had to read it several times). The case of atmospherically isolated water masses applies mainly to deeper layers (below the surface mixed layer, below the euphotic zone) and thus I suggest to use the terms remineralization of organic matter (instead of organic matter cycling) and carbonate dissolution

(instead of carbonate cycling). The results are given in Table 1: the globally most import processes for changes in Ω are cycling of (1) organic matter and (2) carbonate and (3) pressure changes (not really surprising) whereas temperature changes are of minor importance.

When the water sample has the change to equilibrate with the atmosphere (i.e. in the surface ocean), production of organic material has a smaller impact on Ω mainly via the associated change of A_T by nitrate uptake. Freshwater cycling has a large input on A_T and Ω (for the Arctic Ocean compare, for example, Yamamoto-Kawai, Michiyo, et al. "Aragonite undersaturation in the Arctic Ocean: effects of ocean acidification and sea ice melt." Science 326.5956 (2009): 1098-1100.). I don't understand why you discuss pressure changes in this context (surface ocean!),

Process importance (Appendix A): I found it difficult to read Appendix A. If I understand it right the authors calculate the propagation of standard deviations for single processes similar to the standard method known as 'combination of errors' where products of variances and squared partial derivatives are added for various independent variables. The problem here is the complicated dependency of the saturation level Ω on various quantities and processes. In a single process various quantities can co-vary and impact Ω via different 'routes'. The authors try to take these complications into account by summing over seven products of partial derivatives (Eqs. A3 and A5; products stemming from chain rule). In total five different equations are given for the metric M_i. Finally, the authors apply Monte Carlo simulations to estimate variability and uncertainty of the metric (which of the five versions given?).

Throughout the manuscript, the authors talk about 'carbonate cycling' and it remains ambiguous whether they refer to carbonate ions (CO_3^{2-}) or calcium carbonate minerals ($CaCO_3$). Also 'carbonate saturation': replace by carbonate saturation state, introduce Ω which is used later on and give equation.

Further points:

The authors fail to give a thorough introduction into the topic.

p. 11140, L. 20: The tracer Alk* should not be introduced in the introduction, but in the next section "The tracer Alk*".

11140, L. 20:
… to isolate the influences carbonate cycling … ->
… to isolate the influence of carbonate cycling …

p.11141, L. 20: operational definition gives A_T ->
operational definition gives A_T (measured in mol kg^{-1}, gravimetric units)

p. 11141, L. 6-8: This is summary and outlook and is misplaced in the introduction. A proper introduction could include an introduction to the carbonate system, calcium

carbonate minerals, the concept of alkalinity (parts of section 2 could go here), sources of alkalinity (parts of section 3 could go here, p. 11145, lines 8-17) including the role of rivers, and then at the end the main questions posed for this paper.

p.1142 L. 5-6:
… while still mixing and responding to calcium carbonate cycling linearly. ->
… while still mixing linearly and responding to calcium carbonate cycling.

p. 11142, L. 17-19: This sentence is unclear. What is the link between export of organic matter and release of OH⁻? Also, you say a '1:1 release of proton acceptors' - this is a ratio, so 1:1 release per what?.

I assume you refer to an increase of alkalinity by 1 mole when 1 mole of N from nitrate or nitrite is assimilated (Wolf-Gladrow et al., 2007), but please take the time to make the explicit link (from export) to nitrate.

p. 11143, L. 1-5, you're too fast: switch sentences, first introduce empirical value by Kanamori and Ikegami, give equation, then the sentence with the comparison to theoretical value by Wolf-Gladrow et al (give number here, too).

p. 11143, salinity normalization of Robbins (2001). This is a very important part of your Alk* calculation and therefore needs some more explanation besides referring to Robbins. Go step by step, don't mention the subtraction in line 14 yet, that is not relevant here for the definition of what you call 'passive conservative potential alkalinity'. Also give the number for averaged A_p (line 19/20).

p. 11143, L. 20: this is a typical sentence that was written without due diligence: "The mean surface values are chosen to capture the impact of freshwater cycling where precipitation and evaporation occur." After reading the sentence three times, I realized it should read: "The mean surface values were chosen because *we assume that they* capture the impact of freshwater cycling where precipitation and evaporation occur." Sentences like this one are manifold throughout the manuscript.

p. 11144, after eq. 5 add eq. 6 where you give the full equation: Alk* = AT + 1.26 NO_3^- - 66.4 S . Don't add the unit in eq.5 but give it below as: "where Alk* has same units at A_T (µmol kg)."

p.11144 L.8: Mean global surface Alk* is zero by definition, and negative Alk* is possible when potential alkalinity is less than expected from salinity. ->

Mean global surface Alk* is zero by definition, and thus Alk* can take on positive as well as negative values.

p. 11144, l. 14-23: this paragraph comes as a surprise, it is unclear why this is discussed here. This is because the Robbins paper was not thoroughly introduced (see comment above). Maybe it should be in an extra paragraph or section "Evaluation of the tracer Alk*". It definitely needs more explanation of why this is important.

p. 11145, l. 1-17: A_T and Alk* seem to be used intermittently. Do you want to talk about A_T or about Alk*. The part of introducing sources for A_T should be moved to the introduction. Line 14-17: this is disconnected to the previous paragraph, convert to alkalinity units.

p.11146 L.2: The Alk* distribution has a broadly similar explanation to the phosphate distribution. MAY NEED MORE EXPLANATION

p. 11146 L.6: Several qualitative differences between Alk* and phosphate are visible in Figs. 2, 4, and 5. ->
Several differences between Alk* and phosphate patterns are visible in Figs. 2, 4, and 5.

p.11146 L.10: a maxima -> a maximum

p.11147, L1: The nearly-zero mean surface Pacific Alk* indicates that Alk* supply from upwelling and a small riverine source very nearly balances carbonate precipitation. THE FACT THAT THE MEAN SURFACE ALK* OF THE PACIFIC MIGHT IN PART BE DUE TO THE LARGE SURFACE AREA AND THE BY DEFINITION ZERO GLOBAL MEAN OF SURFACE ALK*.

p.11148, L.21: I suggest to drop 'The higher Alk* found for May through July is consistent with Moore et al. (1986)'s radium isotope based finding that 20–34% of the surface waters in this region are derived from Amazon during July vs. 5–9% during December. However, if we assume the Atlantic seawater mixing with the Amazon outflow had an Alk* of 25 −35 μmol kg−1, these Amazon River water fractions would result in Alk* of _−15 to 0 μmol kg−1 in December and 45 to 100 μmol kg−1 in July. We see lower Alk* values in our distribution and a smaller disparity between winter and summer Alk*, suggesting a smaller average Amazon influence for the ocean's surface during both seasons than found by Moore et al. (1986). However, our estimate does not account for any changes in calcium carbonate export induced by nutrient-rich Amazon outflow.' because it leads to nowhere.

p. 11149, l. 4: "intermediate to high": give numbers

p. 11150, L.3: This bay has two high A_T rivers that join and flow into it, the Brahmaputra (1114 μmol kg^{-1}) and the Ganges (1966 μmol kg^{-1}) (Cai et al., 2008).

->

This bay has two high A_T rivers that join and flow into it, the Brahmaputra (A_T = 1114 μmol kg^{-1}) and the Ganges (A_T = 1966 μmol kg^{-1}) (Cai et al., 2008).

or (???)

This bay has two high A_T rivers that join and flow into it, the Brahmaputra (Alk* = 1114 μmol kg^{-1}) and the Ganges (Alk* = 1966 μmol kg^{-1}) (Cai et al., 2008).

Section 3.3: Riverine Alk* regionally: what information is added by this discussion to the study of Cai et al (2008) on alkalinity contribution from rivers? Why is Alk* needed for that discussion?

p. 11153, l. 7-8: what is the outcome of testing this assumption?

p. 11154, l. 2-6: it would be more informative to plot temperature versus surface calcite saturation in a x,y-plot or at least calculate a correlation and give that number.

section 5: Conclusions: not all that is written here is a conclusion by definition, e.g: p. 11154, L. 17-18: "A plot of Alk* against salinity reveals the large A_T input from the Amazon River". That is a sentence for the results section. The conclusion section should be considerably shortened and be reduced to main conclusions. It should include a paragraph on: what are advantages of the Alk* method, why is it needed, what can we use it for that cannot be achieved by A_T alone?

Figures:

The choice of figures seems arbitrary. They are randomly introduced but not discussed at the same time. Page 11145, L. 19-21: First sentence introduces Figure 2. Second sentence introduces Figure 3. There is no discussion of Figure 2. What is the added value of showing the top 50 m mean right after showing the surface values? I can't see any. In line 26 it becomes even clearer: "The subtropical gyres have the lowest open ocean Alk* in Figs. 2, 3 and 4." First, this is badly formulated, but then if you can see the same thing in three figures, two of them are not needed, right?

I suggest to: introduce one figure. Then discuss it. Then introduce the next figure. Then discuss it. Take out the figures where there is no extra information to be discussed.

Same in Figure 6, p. 11146, line 18: In the "2D color histograms" the colors and number of datapoints for certain bins are not further discussed, so the graph could be more simple without colors.

All figures need a revision, just some examples:

* Figure 3: legend for size of dots missing

* Figure 4: the figures look like prepared with ODV. One very helpful feature of ODV is that one can have dots to indicate where samples where taken. This illustrates the density of measurements and gives a feeling of how much one can trust the interpolation. This should be applied here.

* Figure 7: Dots for river outflow are plotted on land.

p. 11156 Eq. (A2) one '∂' missing in eq.

p.11166/7 Tables A1 & A2: Units missing for the partial derivatives (some are dimensionless, however, others possess units).