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Review of ‘A tracer study for the development of in-water monitoring, reporting, and verification (MRV) of ship-based ocean alkalinity enhancement’ by Subhas et al.

The work by Subhas et al. examines the results of a Rhodamine tracer release study conducted near Martha’s Vineyard on the east coast of the United States. The resulting distributions of Rhodamine are used to project the effects of a similar addition of NaOH on carbonate chemistry. This hypothetical alkalinity release experiment is predicted to result in carbon dioxide removal. The ultimate goal of the study is to evaluate the use of the Rhodamine tracer release technique for MRV of ship-based ocean alkalinity enhancement. There is no doubt that this research makes a significant contribution to the growing body of literature on the feasibility of OAE as a marine carbon dioxide removal (mCDR) method. The study is well-designed, the quality of the data and the processing steps are well-documented, and both the data and Matlab code are openly shared and available to readers. In my opinion, this work is worthy of publication after addressing some limitations of the proposed methodology.

We appreciate the Reviewer’s comments and provide some answers and discussion below. Several reviewers raised the issue of OAE and its MRV “research” vs. “deployment”, and we think the distinction here is an important one to make. We will revise the manuscript accordingly, along with addressing the detailed feedback below.

My first comment concerns the calculation of dilution factors. The authors define the dilution factor of Rhodamine using a simple equation (L195), where $D = RT_{in} / RT_{underway}$. This approach does not appear to explicitly account for the diffusion of Rhodamine out of the patch. If the goal is to examine the decay of Rhodamine over time, such a simplification may be sufficient. However, if dilution factors produced in this way are used to estimate the decay of added NaOH, the simplification may lead to inaccurate results. Therefore, it is crucial not to use empirically derived parameterizations without considering the underlying processes.

Dilution is only used in the initial stages during the release itself (approximately 90 minutes), and is used to compare with the semi-empirical ship-wake model of Chou (1996). This equation simply calculates the measured concentration in relation to the initial concentration, and the reviewer is correct that it does not distinguish between how that dilution occurs, i.e. by advection, lateral diffusion, or vertical mixing. Thus, this dilution calculation aligns with the immediate turbulence impact on tracers, similar to how ship wake models parameterize this effect. We plan on adding a short introduction and description of ship-wake models for turbulent dilution in the Methods, and a few sentences summarizing the below content in Section 4.1 of the manuscript.

In the case of dilution, Rhodamine will behave similarly to NaOH. Diffusion, however, depends on the gradient (TA or Rhodamine) between the patch and the surrounding water and could drive NaOH both into and out of the patch, whereas Rhodamine will always diffuse outward, as it is not a naturally occurring tracer. I would argue that diffusion may

play a more significant role—indeed, the decay of Rhodamine in Fig. 6 appears clearly exponential, even on a log scale. While I recognize that the measured range of TA is not large enough to produce drastically different outcomes, it remains important to correctly parameterize the dispersion model. The use of the dynamic baseline seems to improve the results, suggesting that even small variability in TA matters.

As mentioned above, dilution of RWT (and of alkalinity added to seawater) will occur via multiple processes. In all cases, these added materials can only diffuse out of the patch – whereas background constituents and other components of seawater could be exchanged bi-directionally. However, given the timescales and spatial scales above, it is important to distinguish physical mixing and stirring processes, which are often characterized using eddy diffusivities, and will operate on entire water parcels, versus molecular diffusion, which happens on much smaller spatial scales (and much longer timescales).

For instance, lateral eddy diffusivity is on the order of $\sim 5 \text{ m}^2/\text{s}$ (Rypina et al., 2019). Scale analysis of $x^2/t \sim 5$, operating on a patch $\sim 500\text{m}$ in scale such as the one in our experiment, gives a characteristic eddy diffusion timescale t of $500^2/5 = 50,000$ seconds, or 13 hours. Thus, while not important initially, this process clearly controls the mixing and tracer loss on timescales of days. Vertical diffusivity is significantly slower, order $10^{-4} - 10^{-5} \text{ m}^2/\text{s}$, and molecular diffusion is orders of magnitude slower at $\sim 10^{-9} \text{ m}^2/\text{s}$.

We note again, however, that these mixing processes operate on entire water parcels, not on individual components, and it is our hypothesis that eddy-driven mixing and stirring will dilute all components of the patch in a similar manner. It will be critical to carefully evaluate the distribution of RT and NaOH in paired release experiments to test this hypothesis; it will also be useful to confirm that this behavior is captured in ocean models as well. This is our plan for the next field experiment, and for future modeling efforts.

Related to the previous comment, could the authors discuss how the results of the study apply to the use of solid alkalinity sources (e.g., lime, $\text{Mg}(\text{OH})_2$)? I imagine that the proposed framework could be applicable to future deployments, and it is important to understand the limitations of the method.

The advantage of liquid alkalinity is that it can be tracked similarly to the RT dye, whereas solid alkalinity sources pose additional monitoring challenges. Solid -based OAE efficiency will depend on how fast the material dissolves and sinks out of the mixed layer, and both of these processes will decouple the resulting alkalinity enhancement from the dye tracer. We will add these additional considerations – ability to track, dissolution rate, and sinking velocity of solid alkalinity -- in the paper, both in setting up the OAE analytical framework, and in the conclusions.

Finally, I agree with the authors on the benefits of using the dynamic baseline. However, in a larger-scale deployment, identifying or defining "out-of-patch" conditions could be challenging—especially in the absence of a visible tracer. Moreover, once alkalinity

spreads across large areas with varying oceanic conditions and water masses, the concept of “out-of-patch” may lose its relevance. It may be necessary to sample along the patch boundaries, gather several baselines, and integrate them into the proposed framework. A stronger recommendation—which the authors do emphasize in their conclusions—would be to constrain the baseline prior to the experiments by constructing better empirical models (requiring high-resolution data over at least an annual cycle) and characterizing the sources of baseline variability, which could later be incorporated into model-derived counterfactuals.

Thanks for this – we do believe that our sampling plan effectively accomplished this “along-boundary” approach, because we were constantly passing into and out of the patch, thus sampling the boundary multiple times in space and time. Indeed, this approach is what allowed us to construct the dynamic baseline and demonstrate that we were capturing “true” spatial and temporal variability along with any signal of the perturbation itself. We can strengthen this “boundary sampling” approach in the recommendations, as well as understanding how measuring and defining these boundaries might interact with interannual variability.

Minor typos and comments:

- **L51:** Typo – “Research” (please correct). **Done.**
- **L284:** Please verify the sensor models—something seems incorrect. Oxygen is likely SBE 43; please confirm the same for CTD. **Confirmed – thank you for catching this.**
- **L359:** Please use a consistent reference format for Guo et al. throughout the manuscript (e.g., "in review" or "in revision"), unless the paper has already been accepted. Thank you -- This paper has now been accepted with minor revisions and our citation is revised accordingly. If it does become published with a doi we will be sure to update the citation.