

Dear Timothy Bertram,

29 August 2021

Please consider our revised manuscript, “Long-term atmospheric emissions from the Coal Oil Point natural marine hydrocarbon seep field, Offshore, California” for Atmospheric Chemistry Physics Discussion. We have carefully addressed all the issues and constructive critiques of the reviewers, including significant reorganization of the introductory materials and a careful review of the manuscript and supplementary material on a line-by-line basis with numerous edits and rewrites to improve clarity. These little changes were so numerous that track changes became unwieldy, and was not saved, for which we apologize. Responses to reviewers have been uploaded.

One note, in the vast majority of seep field studies, uncertainty values are not reported; however, we feel that by including a thorough uncertainty consideration in our work, we provide an example that hopefully will be repeated within the seep science community.

Data are published at Mendeley Data Repository at doi:10.17632/znhxkftm8.1

Sincerely,

Ira Leifer and co-authors

We would like to take this opportunity to respond to reviewer 2, who both raised some good points, which were addressed through improvements to the manuscript, and some points that do not apply to our study and paper (as written).

**Comment on manuscript length:**

We agree that the introductory and methods section could be organized better, as also commented constructively by reviewer 1, and for which we have shortened, re-organized including removing duplicative and ancillary material. Please see response to reviewer 1. That said, we leave supplemental material as supplemental to (as requested by reviewer 2) not lengthen the manuscript.

**Comment on novelty**

We never argued that the approach is novel - we wrote (and maintain) that it is the application to marine seepage that is novel.

**Comment on THC measurement details missing**

The measurements were made by a regulatory agency and as a result, it should be unnecessary to have the measurement details for the purpose of manuscript evaluation. When modelers use NOAA data they do not report on NOAA measurement approaches. Still, we contacted the agency to find the instrumentation details and added them to the manuscript.

**Comment on uncertainty and sensitivity**

We did an extremely comprehensive series of sensitivity studies because the preferred approach to assess uncertainty – Monte Carlo – is computationally impossible with the resources (workstations) available and an uncertainty approach such as quadrature is inappropriate. Specifically, uncertainty arises from several phenomena for which data are unavailable, such as wind veering and the boundary layer height, and for which both data and processes are unknown such as the emissions ratio between inshore and offshore seepage trends. Quadrature implies that the functional relationship between emissions and the many parameters that affect it can be expressed by an equation – the reality is that for this system, it cannot. The suggestion to run the model with the upper values at  $1\sigma$  is incorrect for assessing uncertainty – it would calculate the upper limit of overall error. To assess uncertainty - a Monte Carlo approach is needed.

We note, that the fact that overall emissions agree reasonably well with published values provides confidence that uncertainty is not so large as to make the model useless.

**Comment on using reanalysis wind and boundary layer product in the inversion**

Our experience of these products is that they fail to capture the necessary fine-scale wind structure on sub-kilometer length scales (clearly evident in the presented data, but perhaps overlooked). In mountainous coastal terrain, such as for California and the study area in particular, Hysplit tends to predict wind trajectories that run right through the coastal mountains. As such these products are inappropriate for the analysis used in this study.

**Comment on not knowing how background THC concentration was derived.**

The derivation of background concentration was provided in the text.

**Comment on potential for biases and errors over time invalidating the effort.**

We did the best we could with available information and data and derived similar values to those reported from snapshot field surveys. Moreover, in the field, annualized budgets are regularly derived from single surveys and assumed to represent emissions even over decades. For example, *Hornafius et al.* (1999) is still cited decades later despite being a 1995 snapshot. Thus, in this regards, an analysis based on 30 years of data obviously improves on current state of knowledge.

Moreover, there is a basis for not expecting dramatic seep field changes on less than geological timescales - geological systems change slowly – with emissions highly spatially constrained by faults and fractures that are “fixed” in solid rock. Specifically, migration from below that charges the shallow reservoir must balance over long times with emissions to the seabed through fractures above. Moreover, as studied in the sensitivity studies, overall emissions are weakly sensitive to shifts in the location of active seepage, which can change on short time scales, even to shifts between the offshore and onshore trends and for along-trend shifts, which were investigated in the wind-veering sensitivity studies.

**Comment on evasion process is complex and not captured by the model.**

We direct the reviewer and reader to the manuscript where we presented data that the highly soluble gas, CO<sub>2</sub>, is in similar ratios in the atmospheric plume and the seabed. This demonstrates that gases that dissolved during bubble rise are transported by the upwelling flow to the sea surface and then evade into the atmosphere. This upwelling process was detailed in the introductory material and in the discussion.

Finally, the reviewer may not have understood how the model functioned. Specifically, overall emissions are not affected whether hydrocarbons evade in the grid cell above the seabed or several grid cells downcurrent – their contribution is counted. Thus, the elevated concentration of seep gas that is observed from a specific direction is assigned to the nearest sources in the sonar map. The model does not consider whether a molecule of methane came from the atmospheric bubble plume or sea surface evasion – a molecule of methane is a molecule of methane – the model simply derives the emissions for the measurement.

Only the fraction that evades from downcurrent of the seep field sonar extent remains unaccounted. This was noted as needed future work.

**Comment on THC not being a conserved tracer.**

Given that the transport time at typical wind speeds is just 20-30 minutes, THC, whose composition was reported, is conserved - the NMHC is predominantly light alkanes with lifespans of weeks to longer. We now note that chemical changes are unlikely on the transport time for completeness.

**Comment that the model assumes constant and uniform emissions.**

Our model does not make this assumption, and thus it is unclear as to how this comment arises.

**Comment that unknown sea-surface state using the partitioning calculate the seabed emissions using a 50:50 partitioning.**

We note that the study goal was to assess the atmospheric emissions NOT to assess seabed emissions. We do so solely to allow comparison with Hornafius et al. (1999). To enable a comparison we used the *Clark et al. (2000)* partitioning, which was based on a large field study. This partitioning is very weakly related to sea-surface state (outside storms) – only to the fraction that dissolves below the wave-mixed layer – dissolved gases above the wave-mixed layer evade sooner (tens of meters) or later (kilometers), and thus were considered by *Clark et al. (2000)* in their partitioning.

**Comment on inability to separate terrestrial and marine sources and emissions**

We agree that for an air quality station in Los Angeles trying to deconvolve the emissions from for example, La Brea, our approach might be ineffective; however, the COP seep field is NOT in Los Angeles. It is offshore. And upwind for prevailing airflow patterns is the vast Pacific. As such the fact that the  $C(\theta)$  distribution closely matches the seep field extent demonstrates the validity of the approach to segregate terrestrial and marine sources – distant sources create very broad features, and present as a gradient across the seep field.

**Comment on the offshore validation study as unconvincing.**

The purpose of the validation study was not to compare with WCS but rather to show that the offshore sources are well-represented by Gaussian plumes, a key assumption.