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Comment on acp-2020-1234

Anonymous Referee #2

Referee comment on "Long-term atmospheric emissions for the Coal Oil Point natural marine hydrocarbon seep field, offshore California" by Ira Leifer et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2020-1234-RC2>, 2021

In this manuscript the authors use a simplified inverse technique to constrain marine emissions of hydrocarbons from a seep area offshore of southern California. The authors draw on a 30-year record of total hydrocarbon concentration (THC) from a ground-based monitoring site to estimate THC emissions from a defined seepage area off the coast of Santa Barbara, California. This estimation is achieved by assuming that THC outgassing is well-described as a Gaussian plume (effectively treating them as point sources). The study region was gridded and each grid was treated as a separate plume. The authors offer a validation of this approach through comparison with shipboard atmospheric measurements from a single day.

Major comments: This manuscript has major flaws. It is overly long, convoluted, and would benefit from a major rewrite, focusing on clarity and brevity. The Introduction contains a large amount of extraneous detail, as do the Methods and Results. Most of the key information on the model is in the Supplemental.

The authors' approach is presented as a novel technique, but upon reading the paper I think it is better described as a crude approximation. There is nothing wrong with simplifying assumptions or simplicity of approach per se, but the authors' description of it as novel is misleading, in my opinion, since there is nothing new about the plume model they employ (they cite a text from the 1980s for the relevant equations). The paper, somewhat incredibly, lacks any details on how the measurements of THC were conducted that I could find. I am not sure, after reading it, what compounds are included in this bulk tracer. Without any information whatsoever it is difficult to properly evaluate the manuscript further.

Another serious shortcoming of the study is the treatment and discussion of uncertainty, which is insufficient. They have done some sensitivity tests of the model, but not attempted to account for, or at least discuss, the uncertainty of their approach. The authors claim that a Monte Carlo simulation was not feasible due to computational constraints. I realize no one has infinite resources, but they could propagate the uncertainty through the model using quadrature sums.

The assumptions underlying the model also require significant justification, which is not presented to my satisfaction. For instance, the authors claim they remove terrestrial

emissions from the model based on wind sector, but this is hardly sufficient--atmospheric transport is complex and this can only account for emissions in the very near field. Why do the authors not use boundary layer height and wind fields from a reanalysis product? There are many such products which are freely available. I am still unclear how they defined a background THC concentration. I do not think the large number of simplifying assumptions made in this approach would be valid over the 30 year record. The potential for biases and errors is too great to make the emission estimates with any confidence. I am doubtful that outgassing relating to hydrocarbon seep ebullition is well-described as a point source. During transport across the sea surface, the hydrocarbons will be diluted and also exchange with the sea surface in vastly different proportions based on their solubilities. Lumping all of these compounds into a single tracer, THC, makes no sense, because it is not a conservative tracer. The plume model assumes that the emission rate is uniform and constant, which is not a valid assumption for seepage ebullition. Even assuming the seepage was completely constant, the state of the sea surface (roughness, surfactants, skin temperature, etc) changes constantly, which will greatly impact the exchange of CO₂, CH₄, HCs, etc. Calculating the seabed emissions from 50:50 partitioning is so crude as to be useless.

A better place to start, in my opinion, would be to calculate a footprint for the WCS station and to convolve this with THC anomalies against a background. Without additional observations of some other conservative tracer for marine air, however, I am still skeptical that they can separate marine emissions from urban/terrestrial emissions. Even in a state of the art inversion, this would be challenging.

The validation study using a field survey is unconvincing, since they are not actually validating through observation that the THC enhancements measured at the station are indeed coming from the seafloor; they are simply comparing additional atmospheric measurements of CO₂, CH₄, and alkanes measured in the atmosphere to the station measurements on a single day. Measurements of dissolved THC would have been more convincing.

The deficiencies here require more than just rewriting or reanalyzing the data, and so I am afraid I cannot recommend publication.