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Comment on acp-2022-452

Anonymous Referee #1

Referee comment on "High accuracy calculation and data quality evaluation of ship emissions based on the sniffer method" by Letian Zhu and Fan Zhou, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-452-RC1, 2022

Review of the manuscript „High accuracy calculation and data quality evaluation of ship emissions based on the sniffer method" by Zhu and Zhou

Shipping emissions are a relevant source of pollution in particular in the clean marine boundary layer. A better characterisation of ship emissions is a relevant topic for atmospheric chemistry studies and for the control of environmental regulations, and improving the accuracy of emission estimates is necessary for a better understanding of the relevance of ship emissions.

In this manuscript, the authors propose a complex algorithm to match air-borne in-situ observations of CO_2 and SO_2 in ship emission plumes with the aim to determine SO_2 emission ratios. Briefly, the algorithm consists of 1) the smoothing of the time series, 2) the assignment of matching peaks in the CO_2 and SO_2 time series 3) the identification and removal of invalid matches and 4) a quality estimate for the remaining matches. The algorithm is described and applied to a small sample of real measurements of variable quality, and good performance of the algorithm is found in comparison to a subjective assignment.

Unfortunately, I cannot recommend this manuscript for publication. The reason for this assessment is that for each of the steps performed, I disagree with the assumptions made and the approaches taken. In many places, ad hoc parameters are introduced and conclusions are drawn without good justification. In addition, the description is hard to follow and often unclear, both because of issues with the use of English and the way the text is written.

If I understood the manuscript well, the authors try to identify the matching maxima in the time series of SO_2 and CO_2 for an individual plume measurement in order to determine the SO_2 / CO_2 ratio from this value. However, I do not see the rationale for using this specific value. The ratio should be constant throughout the plume and therefore could be computed from any pair of matching measurements. If this is not possible for signal to noise reasons, the ratio of the integrals of the two quantities over the plume transect should be taken as this will be less sensitive to noise and to small time lags between the measurements. I therefore think that the whole idea of finding the maxima in the time series is unnecessary and actually not asking the right question.

For the smoothing, the authors try to establish an objective criterion by maximising the "peak density standard deviation". However, while this criterion is quantitative, it is not clear to me why this value should lead to the optimum smoothing.

The next step is the matching of the peaks in the two time series. The authors use a "Dynamic time warping algorithm" with the Manhatten distance in a normalised time-concentration plane to find matching pairs. However, it is not at all clear why a) this distance is a good metric for finding the peaks and b) why taking the absolute values in the Manhatten distance makes more sense than computing the Eularian distance. More importantly, the need for the matching arises from differences in the time response of the SO_2 and CO_2 measurements, which should be constant over the short time periods needed to measure a plume. Therefore, the complex algorithm which assigns peaks in the time series allowing variable time lags results in unphysical assignments. It would have been much simpler and a better representation of the physical effects behind the shift in the time series to calculate cross-correlations of the two time series for different realistic time lags to determine the optimal time shift to be applied to the time series.

As the algorithm proposed in the manuscript is not well constrained (see the last point), it often produces unphysical results. In order to identify and remove them, the authors propose a k-means clustering of the "Normalised Concentration Differences" with two clusters. Why such an approach should be able to separate valid and invalid results is not clear. In my opinion, a simple threshold removing pairs with "too large" differences would lead to similar results and be equally subjective.

The final step in the algorithm is a quality assessment of the derived pairs. Here, the authors apply 16 different index definitions on data sets created by "10000 self-development sampling" and then use a complex scheme to extract a high-level quality index from this set of results. I must admit that I did not follow this indexing in detail, as it appears completely arbitrary and pointless to me.

In summary, I think the manuscript tries to solve the wrong problem with a complex and, in many respects, arbitrary algorithm. Instead, the authors should follow a simple and physics-based approach by comparing the SO_2 and CO_2 values integrated over the plume transect after allowing for a time shift correcting for the different response times of the two instruments used.