An itemized response (blue words) to the reviewer's comments and suggestions

We sincerely appreciate the reviewers for the comments concerning our manuscript entitled "Roles of oceanic ventilation and terrestrial outflow in the atmospheric non-methane hydrocarbons over the Chinese marginal seas" [EGUSPHERE-2023-2935]. These comments are all valuable and very helpful for improving our paper and have an important guiding significance to our future research. We have made changes to the manuscript and tried our best to improve the manuscript following these comments. The changed portion in the revised manuscript is highlighted in blue. The primary corrections in the revised manuscript and the detailed responses to the comments of the reviewer are as follows.

Reviewer #1 Evaluations:

The manuscript is mainly describing the specific effects of ocean emission and terrestrial input on the atmospheric NMHCs in a representative marginal sea. While the impact of terrestrial input on the coastal environment is widely recognized, a more detailed discussion has been lacking. The authors address the influence mechanism of terrestrial land-based inputs on nearshore atmospheric NMHCs, taking into the distance from the shore and relevant parameters of air masses, and assess the relative contributions from land and ocean sources. This is meaningful to deepen the understanding of how terrestrial input affecting the marine environment near the coast. I think the manuscript is suitable for publication in the ACP after a revision.

Reply: We sincerely appreciate the reviewer's valuable comments and suggestions, which help us improve the manuscript. Following the reviewer's comments, we made a revision to the manuscript. All changes in the revised manuscript are highlighted in blue. We also provide below a point-to-point list regarding the changes made in the text for the reviewer's convenience to review.

Major comments or suggestions:

1. The positive matrix factorization model is the methodology employed by the authors to deconvolute the source factors of atmospheric NMHCs and assess their respective contributions. The authors should provide a more detailed exposition on the principles of this approach in Method section to enhance clarity and understanding.

Reply: Thanks for your valuable suggestion. Details for the PMF model have been supplied in the manuscript, as indicated below:

Line 221-245：

"PMF model introduced in detail in the study of Paatero and Trapper (1994) was applied to analyze the data of atmospheric NMHCs in the Yellow Sea and the East China Sea. Based on a matrix consisting of the concentrations of diverse chemical species, the objective of PMF is to determine the number of NMHCs source factors, the chemical composition profile of each factor, and the contribution of each factor to species. The matrix representation of this model is as follows:

$$x_{ij} = \sum_{k=1}^{p} g_{ik} f_{kj} + e_{ij} \qquad (9)$$

Where x_{ij} represents the concentration of species *j* measured on sample *i*, p denotes the number of factors facilitating the samples. f_{kj} represents the concentration of species *j* in factor profile *k*, g_{ik} denotes the relative contribution of factor k to sample *i*, and e_{ij} represents the PMF model error of species *j* measured on sample *i*. The factors resolved by PMF are typically interpreted as sources. The objective of this algorithm is to find the values of f_{kj}, g_{ik}, and p that best reproduce x_{ij}, continuously adjusting f_{kj} and g_{ik} until the minimum Q value for a given p is attained. Q is defined as:

$$Q = \sum_{i=1}^{n} \sum_{j=1}^{m} \left(\frac{e_{ij}}{\sigma_{ij}}\right)^2 \qquad (10)$$

Where σ_{ij} represents the uncertainty of the concentration of the species *j* in sample *i*, n is the number of samples, and m is the number of species. In applying the PMF model, the significance of missing data in the matrix was decreased by using the species median. The uncertainty for normal data was estimated as 20 % of the NMHCs concentrations because the analytical uncertainty was not available (Buzcu and Fraser, 2006). The model ran 20 times and we selected the result with the minimum Q value. Besides, approximately 94 % of the scaled residuals given by PMF ranged from -3 to 3 (Fig. S1), suggesting a reasonable fit of the model result."

2. While the authors have made a clear statement regarding the significant difference in NMHCs between urban and marine atmosphere, there is still a need for a more explicit expression on the distribution characteristics of NMHCs, especially in the marine atmosphere. It is advisable to make advancements in the description within the main text, ideally supplemented with relevant Figure for a comprehensive view.

Reply: Thanks for your valuable suggestion. We have represented an explicit description about the distribution of NMHCs in the marine atmosphere in the main text and provided relevant Figures in supporting information, as indicated below:

Line 271-277:

"In spatial terms, multiple NMHCs (e.g. ethane, propane, i-butane, n-butane, and ethylene) showed higher atmospheric concentrations in regions closer to the land. The elevated concentrations are primarily concentrated along the coastal regions of the East China Sea and the north Yellow Sea (Figure S3). The disparity in NMHCs concentrations between land and ocean, as well as the distribution pattern of NMHCs in the marine atmosphere, suggested the potential influence of terrestrial sources on the oceanic NMHCs."

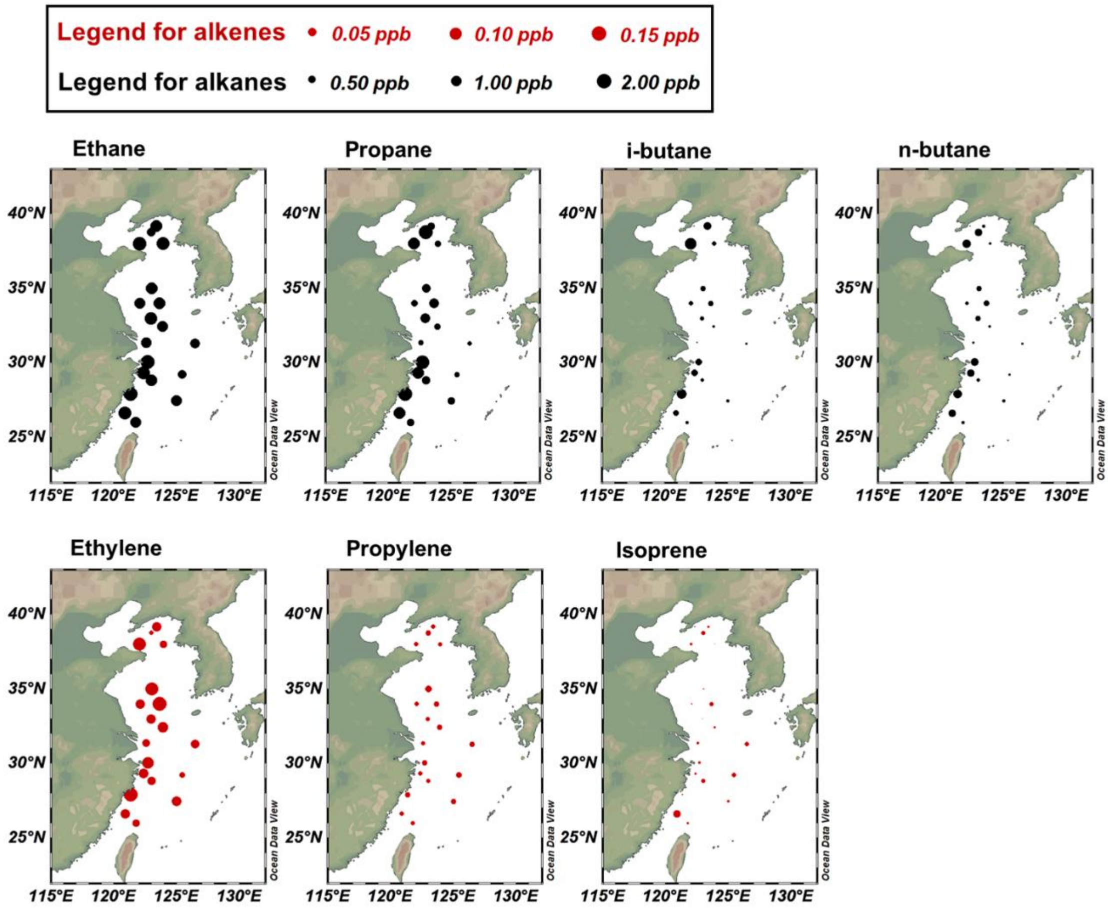

Figure S3 Distributions of alkanes (black dots) and alkenes (red dots) in the atmosphere over the Yellow Sea and the East China Sea

3. Obviously, the distance from the land to the oceanic station is a crucial parameter in the authors' discussion. However, I didn't find any information about the source of distance data or an introduction to the relevant calculation method. The authors should explicitly provide details on these aspects for clarity and transparency in the manuscript.

Reply: We are sorry for our negligence about the introduction of the calculation method for the distance from the land to the oceanic stations. It has been supplied in the manuscript, as indicated below:

Line 196-202:

“2.7 Calculation of the shortest distance from the sampling station to the land

Coastline latitude and longitude data near the study area (20-45°N, 110-130°E) were extracted from the World Vector Shorelines (downloaded from https://www.ngdc.noaa.gov/mgg/shorelines/data/gshhg/latest/). Subsequently, distances from the maritime sampling stations to all coastal locations were computed. The minimum value among these distances was selected as the shortest distance to the land (listed in Table S9).”

4. The authors have employed an innovative approach in assessing the impact of sea-air flux on the marine NMHCs, by calculating the atmospheric lifetime-weighted concentrations of different gases. In this way, the variabilities of atmospheric reactivities of different gases were considered simultaneously when examining the relationship between flux and concentration. It is found intriguing and seemed effective. Furthermore, the authors could extend this novel idea to the discussion about the air mass. Both of land retention and transport time of air masses serve as indicators of air mass characteristics, reflecting the impact of land-based inputs. Combining these two parameters to collectively explore the impact of air masses on NMHCs might offer a new perspective, potentially leading to fresh discoveries

Reply: Thanks for your valuable suggestion. As combining the effects of R_L and transport time of air mass on marine NMHCs, it emphasized the possibility of the terrestrial influence on the marine atmospheric environment.

Line 343-347:

“Notably, elevated alkane concentrations were affected by those air masses with larger R_{L-mean} (>0.8) and shorter transport time (<20 h) (Figure S4). This emphasized the terrestrial influence on alkanes in the marine atmosphere, since both R_L and transport time serve as indicators of air mass terrestrial characteristics.”

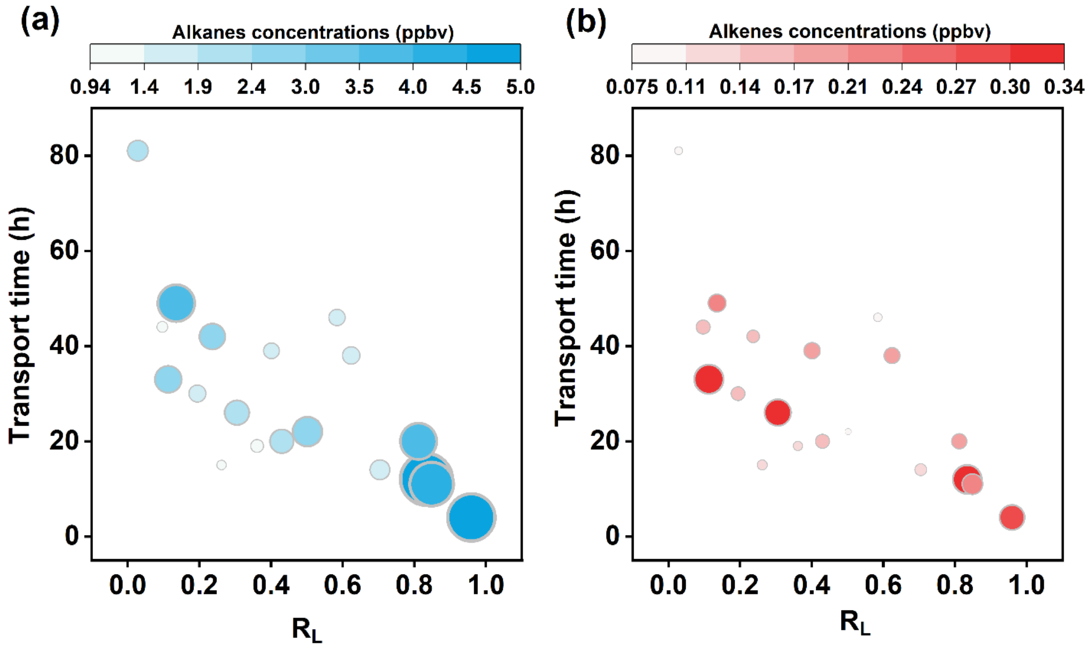

Figure S4 Impacts of air mass (indicated by transport time and R_L) on atmospheric alkanes (a) and alkenes (b) over the Yellow Sea and the East China Sea

Minor comments:

1. Show the standard deviation when you mentioned average, like line 266 “the mean (range) concentration of ethane, propane, i-butane, and n-butane was 2.26 (0.277-5.72), 2.95 (0.149-20.1), 2.57 (BD-27.6), and……”

Reply: Thanks for your valuable suggestion. It has been revised in the manuscript, as indicated below:

Line 251-254:

"In urban atmosphere (n = 14), the mean (range) concentration of ethane, propane, i-butane, and n-butane was 2.26 ± 1.66 (0.277-5.72), 2.95 ± 5.12 (0.149-20.1), 2.57 ± 6.99 (BD-27.6), and 3.29 ± 7.68 (0.018-30.2) ppb, respectively (Table 1)."

Line 257-260:

"For alkene species in the urban atmosphere (n = 14), the mean (range) of ethylene, propylene, and isoprene was 0.180 ± 0.126 (0.035-0.390), 0.036 ± 0.040 (BD-0.129), and 0.046 ± 0.072 (0.006-0.250) ppb, respectively."

Line 262-266:

"In the marine atmosphere (n = 19), the mean (range) concentration of ethane, propane, i-butane, n-butane, ethylene, propylene, and isoprene was 1.24 ± 0.298 (0.686-1.72), 0.822 ± 0.518 (0.226-1.79), 0.283 ± 0.302 (BD-1.17), 0.256 ± 0.214 (0.025-0.694), 0.151 ± 0.077 (0.028-0.295), 0.033 ± 0.009 (0.022-0.060), and 0.008 ± 0.010 (BD-0.043) ppb, respectively."

Line 366-370:

"The mean (range) of sea-to-air fluxes of ethane, propane, i-butane, n-butane, ethylene, propylene, and isoprene was 44.6 ± 35.0 (0.2-118), 41.5 ± 39.9 (0.2-157), 31.7 ± 38.2 (0.1-146), 10.9 ± 25.4 (-0.8-96.1), 321 ±294 (1.7-775), 56.1 ± 55.2 (0.2-212), and 112 ± 134 (0.5-468) nmol m^{-2} d^{-1}, respectively, in the Yellow Sea and the East China Sea (Table 1)."

2. line 358 “ethane possesses an atmospheric lifetime of approximately 78 d at 24 h [•OH] concentration of 6×105 molecules cm-3……”. References are needed to illustrate the source and credibility of the data used here.

Reply: Thanks for your valuable suggestion. We have cited the relevant references as indicated below:

“For instance, ethane possesses an atmospheric lifetime of approximately 78 d at 24 h [•OH] concentration of 6×10^5 molecules cm^{-3} (Jobson et al., 1999), using the rate constant with •OH at 288 K taken from Atkinson et al. (1997).”

3 line 468 “ozone formation potential (OFP) of NMHCs was calculated using OFP = MIR × C…” The calculation description should be in the method section and clearly present the specific constants used in the equation and their literature sources.

Reply: Thanks for your valuable suggestion. The calculation description has been composed in the method section 2.5 and the specific constants have been presented in Table S11.

“5 Calculation of ozone formation potential of NMHCs

$$OFP_i = MIR_i \times C_i \quad (5)$$

To assess the environmental implications of different sources, the ozone formation potential (OFP) of NMHCs was calculated using Eq. (5), where MIR_i depicts the maximum incremental reactivity and C_i represents the concentration of NMHCs (Carter, 1994). Specific data was listed in supplementary Table S11.”

4 Is it necessary to include both the full term and abbreviation 'NMHCs' in the caption of each Figure? Generally, after the initial mention in the text, subsequent references can use the abbreviation alone for conciseness.

Reply: Thanks for your valuable suggestion. After the first appearance of the full name, we use abbreviations in the following text.

5 Increase the font size of the text in the Figures to make them more readable.

Reply: We feel sorry for the blurred figures due to the inappropriate font size. We have increased the font size of the text in the Figures, e.g. Figure 7.

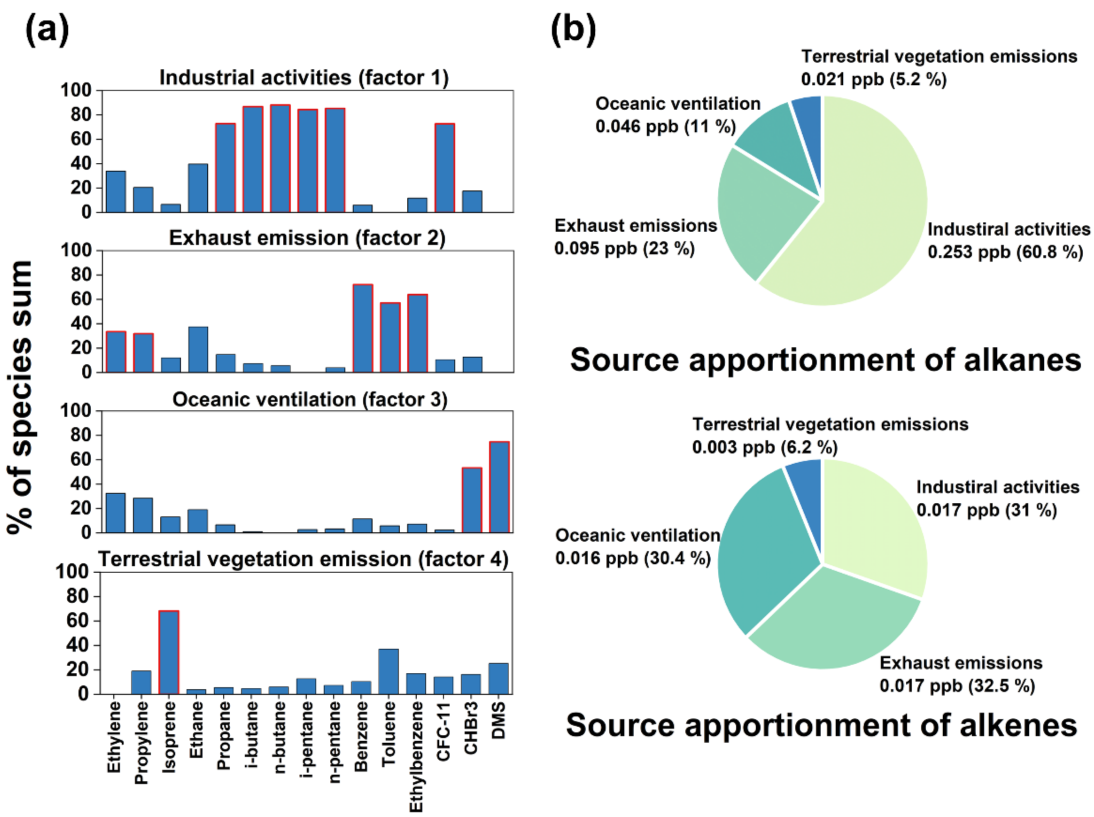

Figure 7 Representative factor profiles from the PMF model (panel a) and relative contributions of different factors/sources to the alkanes and alkenes in the oceanic atmosphere (panel b). NMHCs in

panel a marked with red rim are selected as indicators for the specific factors.