Dear Editors and Reviewers,

Thank you for your valuable comments on our manuscript entitled "Modeling of PAHs From Global to Regional Scales: Model Development and Investigation of Health Risks from 2013 to 2018 in China" (MS No.: EGUSPHERE-2024-1437). Those comments are greatly helpful for improving our manuscript. We provided point-by-point replies to your comments and revised the manuscript accordingly (in **blue and highlighted**).

The comments (in black) are copied here and responses to the comments (in blue) are as follows:

Response to Referee #2:

The study investigates the global distribution of polycyclic aromatic hydrocarbons (PAHs) and their health risks, focusing on China from 2013 to 2018. Using the IAP-AACM model, the authors evaluate the spatial and seasonal distribution of BaP, a key PAH indicator. The authors show BaP concentrations have decreased in China due to emission reductions but increased in India and Southern Africa. Despite the reductions, the health risks in China, particularly in East China, remain significant.

By developing a PAH module, the study offers significant insights into PAH distribution and health risks, providing valuable insights into PAH trends across developing regions. I am happy to see its publication in due course. I would like to suggest some minor revisions after addressing the following questions:

Response: Thank you so much for your constructive comments and insightful suggestions. We have revised the paper carefully.

<u>Comment 1:</u> The authors should justify the capability of the EDGAR inventory to capture the short-term emission changes between 2013-2018 at the regional scale. <u>Response:</u> Thank you for your valuable comments. The emission inventories of BaP in this paper were derived from the Emissions Database for Global Atmospheric Research (EDGAR) and Peking University (PKU). According to statistical data, China was one of the largest PAH-emitting countries in both inventories, accounting for about 27.2% and 29.6% of the world emission in 2013, respectively. Africa and South Asia had the second and third-largest emissions, with residential combustion accounting for 87.3% and 84.0% of total emissions in 2013, respectively. This is related to the widespread use of biomass fuels for heating and cooking in developing countries.

We further calculated the changes in BaP emissions between 2013 and 2018 based on EDGAR inventory and found that BaP emission in China decreased by 10.4%, which is consistent with Wang et al. (2021) (PAH emissions decreased by 11.36% from 2013 to 2017). The results of the change in emissions from different sectors showed that the industrial sector contributed the most to the decline of BaP emissions, followed by the residential combustion, which decreased by 18.9% and 5.1% from 2013 to 2018, respectively, (Figure R1), Wang et al. (2021) also showed that emissions from the industrial and residential/commercial sectors declined by 17.32% and 10.58% from 2013 to 2017.

The simulated concentration and their change are also an indication of the emissions as the concentration is largely determined by emission. Compared to 2013, the simulated concentrations of BaP in 2018 showed a decreasing trend in Germany, Poland, Czech Republic (in Europe), Chicago, Sturgeon Point, and Cleveland (in United States), which is the same as the results from observational datasets (EMEP and IADN). In contrast, the concentrations of BaP in developing countries such as Africa and India showed an increasing trend, which is related to high population growth rates lead to increased residential combustion and energy consumption activities in developing countries (Han et al., 2022). In addition, the PKU emission data showed a similar change in BaP emission. Overall, the EDGAR inventory has a reasonable emissions distribution and the capacity to capture short-term emissions changes relatively well. According to your suggestions, we have added a figure for emissions and changes in some regions and relevant descriptions in the supplement and revised manuscript to show the ability of EDGAR inventory to capture the short-term emission changes. (Line 265-267, 269-274, and 279-286)



Figure R1. Emissions and changes for different sectors in China, Africa, South Asia, Europe, North America, and Sorth America in 2013 and 2018. This figure corresponds to Figure S3 in the revised manuscript.

<u>Comment 2:</u> Figure 8 looks like just a zooming in of Figure 4 with a focus over China. Again, how to verify the very local changes in Figure 8?

<u>Response</u>: We apologize for the lack of clarity giving rise to misunderstanding. Figure 8 is not just a zooming-in of Fig. 4. Actually, we used a multi-scale domain-nesting approach to simulate BaP over China. Figure 4 shows the global spatial distributions of annual mean BaP concentrations at low resolution $(1^{\circ} \times 1^{\circ})$, and Fig. 8 shows the regional distribution at fine resolution $(0.33^{\circ} \times 0.33^{\circ})$. Figure R2 showed the absolute (Fig. R2a and 2c) and relative concentration changes (Fig. R2b and 2d) at different resolutions in China. According to your suggestion, we have revised the colorbar of Fig. 4c-f to be consistent with Fig. 8, making the difference more visible.

Furthermore, we agree with you that it is important to verify the regional changes. The results of the model are affected by uncertainties from the emission inventory, and yet the observations in China are very sparse and most of the data are not continuous in time, which makes it very difficult to validate the results of the changes in China. Compared with the references (Line 447-448 and 452-454), we found that the simulated concentration of BaP reproduced the declining trend in Beijing (Lin et al., 2024), Shanghai (Yang et al., 2021), Shenyang (Zhang et al., 2023), Tianjin (Zhang et al., 2022), and Chengdu (Xue et al., 2024). The concentration of BaP showed an increase in the Sichuan basin when meteorological conditions were considered. This may be related to the decrease in temperature and planetary boundary layer height compared to 2013, which is unfavorable for pollutant diffusion (Ding et al., 2019). In summary, our simulation results can basically reflect the regional variations of the BaP concentrations.



Figure R2. The (a/c) absolute and (b/d) relative concentration changes (b/d) of annual mean BaP concentrations (a, b) at low resolution and (c, d) fine-resolution in China are shown, respectively.

<u>Comment 3</u>: In Figures10-12, it is not easy for the readers to understand the abbreviation of each province.

Response: We completely agree with this point. It is a bit difficult and inconvenient for the reader to understand abbreviations for each province in Fig. 10-12. As shown in the Figure below, we have used the full name of the province instead of the abbreviation in

Fig. 10-12 and deleted the text "the provinces are listed in Table S4)". In addition, Fig.S4-S6 has also been modified in the supplement.



Figure R3. The TILCR values for the three age groups (Children, Women, and Men) in different provinces of China in 2013. This figure corresponds to Figure 11 in the revised manuscript.

<u>Comment 4</u>: I am wondering what's the motivation to focus on the PAH health risks over China, and it is possible to include more developing regions?

Response: In the paper, we mainly focused on the PAHs health risks in China for two reasons: Firstly, as air quality in China has been improved dramatically, it is necessary to assess the impact of "the Action Plan on Air Pollution Prevention and Control" promulgated by the State Council of China in 2013 on the concentration of BaP in the atmosphere and the health risks. Secondly, according to Eq. (14)-Eq. (18), the health risk is positively correlated with the concentration of BaP in the atmosphere. As can be

seen from the spatial distribution of annual mean BaP concentrations (Fig. 4 in the manuscript), the BaP concentrations in China are significantly higher than those in other developing regions.

Thanks for your good suggestion from a global perspective, we have also added a figure (Fig. 7) and the description of the global distribution of health risks in Sect.4.1 in the revised manuscript (Line385-398). As can be seen in Figure R4, the most of the countries (except China) just face negligible cancer risk, and no regions facing high potential cancer risk when evaluation is based on annual mean concentration. However, it should be noted that TILCR values increased in 2018 compared to 2013. For example, the highest TILCR in Africa in 2018 is twice as high as 2013 and changed from negligible to potential cancer risk. These countries will be likely to face potential even high potential cancer risks, especially in winter. In addition, Lou et al. (2023) has indicated that the peaks of health risks will likely shift from East Asia in 2008 to South Asia and Africa by 2050, due to the increase of residential fuels use along with the population growth in these regions. Therefore, it is clear that clean development is a necessary consideration for developing countries to avoid the health risks posed by PAHs.



Figure R4. The distribution of health risks grade in (a) 2013 and (b) 2018, the distribution of TILCR in (c) 2013 and (d) 2018, and the absolute from 2013 to 2018 when considering the change in (e) emissions only, (f) both emissions and meteorological conditions. This figure corresponds to Figure 7 in the revised manuscript.

<u>Comment 5:</u> In terms of PAH control, I suggest to add more policy-related discussions. For example, more specific measures could be proposed.

Response: This is a very good point. The policy-related discussions are added and specific measures are proposed for PAH control based on our analysis (Line 89-92, 411-412, 469-470, and 596-601).

Line 89-92: "This Action Plan established many effective emission reduction and energy-saving policies, such as strengthening industrial emission standards, eliminating outdated polluting industries, upgrading industrial boilers, and developing clean fuels in the residential sector".

Line 411-412: "This can be largely attributed to the industrial and residential coal combustion in these regions".

Line 469-470: "The concentration also decreased in the North China Plain, benefitting from the emission reduction policies such as the development of cleaner energy sources and the control of industrial emissions".

Line 596-601: "Especially in the fall and winter seasons, the concentration of BaP and the associated health risk is significantly higher than in other seasons. Management efforts on key sectors (e.g., industrial and residential sources) should be further strengthened. In heavily polluted cities, using clean energy to replace coal combustion, adjusting the energy structure of factory production, and developing innovative technologies with lower and even no emissions would be helpful to reduce PAHs pollution".

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