

In this study, PAHs in atmosphere were analyzed from 2014 to 2019 in the Bohai Sea in China. It was found that the atmospheric concentrations were decreased in this period, and the composition of PAHs was also changed caused by the pollution prevention and control. Finally, the cost with the lung cancer caused by PAHs exposure was also estimated, and it was also found the decreasing trend from 2014 to 2019. In general, the study was well organized, and the data was enough for obtaining strong conclusion as the study mentioned. The study can be considered as a key area for the air pollution control in China, which also has scientific contributions to related field. The structure and English writing were easy for understanding. There are some suggestions for the revision.

(1) Some writings should be carefully checked, such as BS is the most polluted area of PAHs in China? Is there any supporting information?

Thank you very much for the advice. The Bohai Rim region was the third pole of China's economic development, with many economic development areas. The rapid population growth, urbanization, and industrialization led to increased pollution for the BS region. It was estimated that the PAHs emissions of the three provinces around the Bohai Sea Economic Circle (Liaoning, Hebei, and Shandong) accounted for 21% of the national emissions (Shen et al., 2013). And the emissions of the near four provinces (Shanxi, Henan, Anhui, and Jiangsu) accounted for 20% of the national emissions (Xu et al., 2006). Under the effect of the west wind, the outflow to the east accounted for 80% of the total outflow, and the emission source was mainly in North China (Zhang et al., 2011). In the process of atmospheric outflow, about 70% of PAHs deposited at the coastal zone and offshore area in the east of China (Lang et al., 2008). Therefore, the concentration of PAHs at the BS and its surrounding areas was at a relatively high level. Therefore, the original sentence of the manuscript was changed to “The Bohai Sea (BS) as one of the severe polluted areas of polycyclic aromatic hydrocarbons (PAHs) in China has been received wide attention in recent decades.”, see Line 22-23 and “PAHs pollution in the atmosphere of the Bohai Sea was in a severe situation (Wang et al., 2018).”, see Line 65-66.

(2) In the introduction section, the relationship between BTH and BS should be clearly mentioned. In my opinion, too many information was added for BTH, however, more information should be added for BS.

Thanks for the advice. The paper added the introduction of the relationship between the BTH region and the BS region. “The Bohai Rim economic area included the Liaodong Peninsula, the Shandong Peninsula, and the Beijing-Tianjin-Hebei (BTH) region. The BTH region was the center of economic development of the Bohai Rim economic area. (Liang et al., 2018; Zhang et al., 2016)”, see Line 66-68.

(3) Line 125, what’s the objective of adding hexamethylbenzene?

Hexamethylbenzene was used as an internal standard for the instrumental quantitative analysis.

(4) For QA and QC, the information for background was missing, which was important for PAHs analysis.

Each batch of samples contained one laboratory blank and one field blank. The pretreatment process of the blanks was consistent with the samples. No target of PAHs was detected in laboratory blanks. Acy, Ace, Flu, Phe, Fla, and Pyr were detected in field blanks, and the values of Acy, Ace, Flu, Phe, Fla, and Pyr were 0.04 - 0.10, 0.04 - 0.09, 0.03 - 0.08, 0.01 - 0.04, ND (Not detected) - 0.01, and ND (Not detected) - 0.02. Relevant information was added to the Table S2 of SI.

(5) For the calculation of ILCR, is the method widely applied in related studies? As I know, the USEPA has other method for ILCR with more parameters, such as body weight, breathing-rate.

Thank you very much for the suggestion. The USEPA has the method for ILCR with more parameters, such as body weight, breathing-rate. It could also be estimated by using Eq. (4), and it had been approved (Zhuo et al., 2017).

(6) Lin 213 to 218, if the cited studies did not use the passive air sampling method, the comparison cannot be made. Because there are some uncertainties between active and passive air sampling methods, which will influence the comparison conclusion.

Thank you very much for your advice. There was some uncertainty between active and passive air sampling methods. In this study, the sampling volume of the passive sampler

was taken into consideration when calculating the PAHs concentration of the sample, which has been reported in previous studies (Jaward et al., 2005; Moeckel et al., 2009).

(7) Line 300, for figure 2 the error bar should be added for average or mean values, the same problem should also be corrected for the whole manuscript.

The error bar in Figure 2 was added, see Line 310. Other relevant issues have been corrected, such as Figure 5 shown in Line 435.

(8) For Section 3.2, TJ was selected as the special site for BS, and the sources apportionments were both analyzed for TJ and BS. I just want to know, for the analysis of BS, the data of TJ was excluded from BS or not? If the data of TJ was included in BS for analysis, it may influence the results, please check.

Thank you for your advice. The data of TJ was not excluded from the BS. Since this study evaluated the source apportionment was analyzed for the entire BS, TJ was not separated from the BS. At the same time, TJ was an important PAHs emission source for the BS. (Zhang et al, 2016)

(9) Line 328, what's the type of fuel combustion?

Line 328 "Fuel combustion emissions were the reason for the significant increase of atmospheric pollutants, and that were also responsible for the elevated PM_{2.5} levels around the BS region." in the sentence "Fuel combustion" referred to the combustion of fossil fuels, and it has been modified in the manuscript, see Line 339.

(10) Line 383-384, the related figure should be added, just like Figure 4.

Line 383-384 "Generally, the seasonal distribution of five sources for atmospheric PAHs at TJ was consistent with that the BS, which was not separately discussed here". The relevant information of TJ was shown as Figure S10 of SI, see Line 395-396.

(11) Line 411-412, the sentence was confused.

Thank you for your suggestion. "The decrease in the fifth year compared with the first year was more than two times, indicating that the environmental health risk of PAHs was decreasing" was modified to "The environmental health risk of PAHs in the fifth year was decreased by three times than in the first year", see Line 421-423.

(12) Line 436, the value of ILCR was higher than 10^{-4} , which indicated life-threatening for human Same as Comment (5), for Section 3.3, the major concern is the suitability

of Eq. (4), please check.

Firstly, Line 436 meant to express the whole ILCR level of the Bohai Sea. Eq. (4) in the reference research (Zhuo et al., 2017), it referred to “Health risk due to inhalation exposure of PAHs was characterized by estimating the Incremental Lifetime Cancer Risk (ILCR) using a point-estimate approach”. The sites set up in this study involve the whole Bohai Sea region and were evenly distributed. The evaluation of ILCR around the Bohai Sea was based on the overall evaluation of 12 sites atmospheric PAHs concentrations.

(13) Line 484, the two scenarios should be mentioned clearly here.

Thank you very much for the advice. Corrections have been made in the manuscript, the “Figure 6 shows the comparative results of direct medical costs of lung cancer at the BS region and TJ under two scenarios from 2014 to 2018” was changed into “Figure 6 shows the comparative results of direct medical costs of lung cancer at the BS region and TJ from 2014 to 2018 under before and after pollution control”, see Line 493-494.

(14) Line 488, five times? Please check.

Thank you for your advice. Modifications have been made see Line 492 “Assuming that no air pollution control was implemented, the total direct medical costs of lung cancer caused by PAHs exposure did not change in five years, and the total direct medical costs was \$23.2 million.”, see Line 498.

References

Liang, X., Tian, C., Zong, Z., Wang, X., Jiang, W., Chen, Y., Ma, J., Luo, Y., Li, J., Zhang, G.: Flux and source-sink relationship of heavy metals and arsenic in the Bohai Sea, China, *Environ. Pollut.*, 242, 1353-1361, <https://doi.org/10.1016/j.envpol.2018.08.011>, 2018.

Zhang, Y.J., Lin, Y., Cai, J., Liu, Y., Hong, L.N., Qin, M.M., Zhao, Y.F., Ma, J., Wang, X.S., Zhu, T., Qiu, X.H., and Zheng, M.: Atmospheric PAHs in North China: Spatial distribution and sources, *Sci. Total. Environ.*, 565, 994-1000, <https://doi.org/10.1016/j.scitotenv.2016.05.104>, 2016.

Shen, H., Huang, Y., Wang, R., Zhu, D., Li, W., Shen, G., Wang, B., Zhang, Y., Chen,

Y., Lu, Y., Chen, H., Li, T., Sun, K., Li, B., Liu, W., Liu, J., Tao, S.: Global Atmospheric Emissions of Polycyclic Aromatic Hydrocarbons from 1960 to 2008 and Future Predictions, *Environ. Sci. Technol.*, 47, 6415-6424, <https://doi.org/10.1021/es400857z>, 2013.

Xu, S.S., Liu, W.X., Tao, S.: Emission of polycyclic aromatic hydrocarbons in China, *Environ. Sci. Technol.*, 40, 702-708, <https://doi.org/10.1021/es0517062>, 2006.

Zhang, Y., Shen, H., Tao, S., Ma, J.: Modeling the atmospheric transport and outflow of polycyclic aromatic hydrocarbons emitted from China, *Atmos. Environ.*, 45, 2820-2827, <https://doi.org/10.1016/j.atmosenv.2011.03.006>, 2011.

Lang, C., Tao, S., Liu, W., Zhang, Y., Simonich, S.: Atmospheric transport and outflow of polycyclic aromatic hydrocarbons from China, *Environ. Sci. Technol.*, 42, 5196-5201, <https://doi.org/10.1021/es800453n>, 2008.

Zhuo, S., Shen, G., Zhu, Y., Du, W., Pan, X., Li, T., Han, Y., Li, B., Liu, J., Cheng, H., Xing, B., and Tao, S.: Source-oriented risk assessment of inhalation exposure to ambient polycyclic aromatic hydrocarbons and contributions of non-priority isomers in urban Nanjing, a megacity located in Yangtze River Delta, China, *Environ. Pollut.*, 224, 796–809, <https://doi.org/10.1016/j.envpol.2017.01.039>, 2017.

Jaward, T.M., Zhang, G., Nam, J.J., Sweetman, A.J., Obbard, J.P., Kobara, Y., and Jones, K.C.: Passive air sampling of polychlorinated biphenyls, organochlorine compounds, and polybrominated diphenyl ethers across Asia, *Environ. Sci. Technol.*, 39, 8638–8645, <https://doi.org/10.1021/es051382h>, 2005.

Moeckel, C., Harner, T., Nizzetto, L., Strandberg, B., Lindroth, A., and Jones, K.C.: Use of de-puration compounds in passive air samplers: results from active sampling-supported field deployment, potential uses, and recommendations, *Environ. Sci. Technol.*, 43, 3227–3232, <https://doi.org/10.1021/es802897x>, 2009.