Response to the comments on the manuscript (HESS-2024-387) **"Mapping mining-affected water pollution in China: Status, patterns, risks, and implications"** by Ziyue Yin, Jian Song, Dianguang Liu, Jianfeng Wu*, Yun Yang, Yuanyuan Sun, and Jichun Wu.

Note that the following text in Arial Narrow font denotes Referee's comments and in Times New Roman font denotes our response to the comments in the review. In our resubmission, the marked PDF file (HESS-2024-387_R1_marked.pdf and Supplement_R1_marked.pdf) has clearly indicated all changes to the original manuscript, tables and figures. Also, in our marked PDF file, marked in ~~a green strikethrough font~~ is the text that should be removed from the original manuscript and marked in a red font is the text that has been added to the current revision. In addition, Line number(s) mentioned below can be referred to as that line numbering in the marked revised manuscript.

Response to Referee #3's Comments

The study delivers a thorough and spatially explicit examination of mining-induced water pollution and associated health risks across China, utilizing an extensive dataset comprising 8,433 samples. The differentiation between coal and metal mines, along with the identification of southern China as a pollution hotspot, provides valuable insights for region-specific policy formulation. Below are several constructive suggestions for refining the manuscript:

[Response] We sincerely appreciate your constructive comments and the recommendation for minor revisions of the manuscript. Moreover, we have made the necessary changes to the original manuscript and hereinafter provided a point-by-point response.

1. While the spatial heterogeneity of pollution is convincingly presented, the underlying mechanisms driving the pronounced contamination in southern China (e.g., geological factors, mining practices, or climatic conditions) warrant further elaboration. Incorporating a brief discussion that connects regional geochemistry or historical mining activities to observed pollution patterns would enhance the robustness of the analysis.

[Response] Thank you for your positive and constructive suggestions. As you suggested, a further elaboration of the underlying mechanisms (*e.g.*, geological factors, mining practices, and climatic conditions) driving the pronounced contamination in South China has been added in Section 4.1 of

the revised manuscript. Furthermore, to enhance the robustness of the analysis, the potential relation between the observed water pollution patterns and both regional geochemical characteristics and historical mining activities has been discussed in detail (**Lines 534-573**):

The underlying mechanisms, including climatic conditions, geological factors, and mining practices, determine the spatial patterns of mining-affected water pollution in China, especially in the highly polluted southern regions. In terms of climatic conditions, the average temperature of the coldest month is > 0°C, while that of the hottest month is > 22°C, and the annual average precipitation is generally > 1,000 mm in South China. The high temperature and precipitation create a synergistic accelerator for mine water acidification. Elevated temperatures stimulate acidophilic microbial communities (*e.g.*, *Acidithiobacillus ferrooxidans*), which enhance enzymatic activity that catalyzes sulfide mineral oxidation. Combined with high levels of precipitation, rainfall infiltrates abandoned mines, tailings ponds, and exposed ore bodies, creating a sustained water-oxygen exchange that drives sulfuric acid formation and iron oxidation processes.

In terms of geological factors, the unique geo-environmental settings of South China, characterized by rugged topography, widespread sulfur-rich strata, and high background value of metallic minerals, result in mining-affected water with high acidity and elevated concentrations of sulfate, Fe, Mn, and HMs (Sun et al., 2022). The coal-forming periods of different mines in the South China coalfields are diverse, mainly Triassic, Neoproterozoic, etc., of which the sulfur enrichment exhibits strong links to marine-land interactions. The sustained seawater intrusion-regression cycle results in elevated sulfur contents (predominantly medium and high-sulfur coals) (Ai et al., 2023; Sun et al., 2025). As illustrated in Fig. S2, the coal fields in China exhibit sulfur contents ranging from 0.02% to 10.48%, with South China's coal-bearing areas showing the highest weighted average sulfur content (2.35%), including 29.63% of high-sulfur coal. Comparatively, those weighted average sulfur contents of coal-bearing areas in Tibet-Western Yunnan, North China, and Northeast China are 0.94%, 0.88%, and 0.86%, respectively (Tang et al., 2015). In addition, as shown in Fig. S3, the metal mineral resources are abundant in the southern region of China, and the water affected by mining practices is often highly toxic, with harmful HMs such as Cd, Pb, Hg, Cr, As, Cu, and so on, endangering the surface water and groundwater systems (Sun et al., 2022).

As to mining practices, especially those involving sulfide-bearing metalliferous ore deposits and sulfide-rich coal mining, are intrinsically associated with AMD. Acid drainage can occur wherever sulfide minerals are excavated and exposed to atmospheric oxygen. The main sulfide minerals in mine wastes are pyrite (FeS_2) and pyrrhotite ($Fe_{1-x}S$), while other associated sulfides are prone to oxidation and release toxic elements, including Al, As, Cd, Co, Cu, Hg, Ni, Pb and Zn, into the water flowing through the mine tailings (Blowes et al., 2014). The oxidation of FeS_2 by

atmospheric oxygen can be expressed by Eqs. (R1) - (R4). Moreover, underground mining is the primary exploitation method in China. Substantial mined-out areas are formed after mining activities, inducing the accumulation of groundwater and the formation of acid mine water. In recent years, the phenomenon has intensified because a number of mines are abandoned without proper closure measures (Jiang et al., 2020).

$$2FeS_2 + 7O_2 + 2H_2O \rightarrow 2Fe^{2+} + 4SO_4^{2-} + 4H^+ \quad (R1)$$

$$4Fe^{2+} + O_2 + 4H^+ \rightarrow 4Fe^{3+} + 2H_2O \quad (R2)$$

$$Fe^{3+} + 3H_2O \rightarrow Fe(OH)_3 + 3H^+ \quad (R3)$$

$$FeS_2 + 14Fe^{3+} + 8H_2O \rightarrow 15Fe^{2+} + 2SO_4^{2-} + 16H^+ \quad (R4)$$

Ai Y.L., Chen, H.P., Chen, M.F., Huang, Y., Han, Z.T., Liu, G., Gao, X.B., Yang, L.H., Zhang, W.Y., Jia, Y.F., and Li, J.: Characteristics and treatment technologies for acid mine drainage from abandoned coal mines in major coal-producing countries. J. China Coal Soc., 48(12), 4521-4535 (in Chinese with English abstract), https://doi.org/10.13225/j.cnki.jccs.2022.1846, 2023.

Blowes, D.W., Ptacek, C.J., Jambor, J.L., Weisener, C.G., Paktunc, D., Gould W.D., and Johnson, D.B. The geochemistry of acid mine drainage. Treatise on Geochemistry (Second Edition), 11, 131-190, https://doi.org/10.1016/B978-0-08-095975-7.00905-0, 2014.

Jiang, C.F., Gao, X.B., Hou, B.J., Zhang, S.T., Zhang, J.Y., Li, C.C., and Wang, W.Z.: Occurrence and environmental impact of coal mine goaf water in karst areas in China. J. Cleaner Product., 275, 123813, https://doi.org/10.1016/j.jclepro.2020.123813, 2020.

Sun, Y.J., Guo, J., Xu, Z.M., Zhang, L., Chen, G., Xiong, X.F., Hua, J.F., Mu, L.J., and Wu, W.X.: Spatial distribution characteristics of mine water quality in coal mining areas of China and technological approaches for mine water treatment. J. China Coal Soc., 50(1), 584-599 (in Chinese with English abstract), https://doi.org/10.13225/j.cnki.jccs.YG24.1547, 2025.

Sun, Y.J., Zhang, L., Xu, Z.M., Chen, G., Zhao, X.M., Li, X., Gao, Y.T., Zhang, S.G., and Zhu, L.L.: Multi-field action mechanism and research progress of coal mine water quality formation and evolution. J. China Coal Soc., 47(1), 423-437 (in Chinese with English abstract), https://doi.org/10.13225/j.cnki.jccs.YG21.1937, 2022.

Tang, Y.G., He, X., Cheng, A.G., Li, W.W., Deng, X.J., Wei, Q., and Li, L.: Occurrence and sedimentary control of sulfur in coals of China. J. China Coal Soc., 40(9), 1977-1988 (in Chinese with English abstract), https://doi.org/10.13225/j.cnki.jccs.2015.0434, 2015.

2. The health risk assessment (e.g., 51.52% carcinogenic risk for adults) raises significant concerns but lacks sufficient methodological detail. Please specify the exposure parameters employed (e.g., ingestion rates,

body weight assumptions) and the toxicity thresholds applied. Additionally, clarify whether the identified risks stem from specific contaminants (e.g., arsenic, cadmium) or synergistic interactions among multiple pollutants.

[Response] Comment accepted. The main parameters used for the human health risk assessment (*e.g.*, ingestion rates, exposure frequency, body weight, etc.) are presented in Table S2. The values of permeability coefficient of skin (K_p), reference dose (RfD_o), gastrointestinal digestion coefficient (ABS_{GI}), and slope factor (SF) for each element are described in Table S3 in Section S3 of the **Supplement**. In the study, Cr, Cd, and As are considered for the calculation of the carcinogenic risks. On the other hand, Fe, Mn, Cr, Ni, Cu, Zn, As, Cd, and Pb are taken into consideration to identify the non-carcinogenic risks. Note that the identified risks (*e.g.*, 51.52% carcinogenic risk for adults) stem from synergistic interactions among multiple pollutants, which have been clarified in the revised manuscript to avoid unnecessary misunderstandings (**Lines 479-482** and **Lines 517-520**).

3. The temporal dimension of water sampling remains ambiguous. Were the samples collected across different seasons or years? Temporal variability in water chemistry (e.g., the influence of monsoon events on metal mobility) could significantly affect risk estimates and merits further exploration in the discussion section.

[Response] We appreciate your insight. Indeed, the dataset we used in the study was collected from the published literature over the past decades, and the surface water or groundwater samples were collected from different years (1964 ~ 2024) and seasons/months. To address your concerns and improve the readability of the revised manuscript, the detailed temporal dimension of the sampling year and sampling month has been supplemented in Table S1 in the ESM2.xlsx. Based on your suggestions, we have briefly explored the impact of temporal variability in water chemistry on risk assessment in the **Discussion** of the revised manuscript (**Lines 718-725**):

Temporal variations in water chemistry (e.g., seasonal fluctuations and monsoon events) significantly impact the environmental fate of contaminants and health risks through multiple mechanisms. During the monsoon season, heavy rainfall flushes tailings ponds or open-pit mines, causing instantaneous spikes in HMs (e.g., Cd, Cr, and As) and sulfate concentrations. Meanwhile, the elevated groundwater levels associated with high precipitation infiltration drive contaminant plumes along preferential pathways. These dynamics introduce systematic biases into traditional static risk assessments. The annual or quarterly average risk assessment model may underestimate short-term high-dose exposure risks. However, the temporal dimension of the dataset used in the study is not yet sufficient to further explore the above issues from a national-scale perspective. Therefore, we will provide an in-depth insight into our future studies.