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\_ATC1.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 #1's Comments**

Human activities of mining have profound impacts on water quality at local to regional scales. In this paper, the attention is paid to mapping mining-affected water pollution in China. It is achieved through the compilation of a number of publicly available surface water and groundwater datasets. In general, the paper can be valuable.

[Response] No change needed. We sincerely appreciate your positive and insightful comments. Hereby we have fully addressed all of your concerns into the revised manuscript and made the necessary clarifications below.

There are three major comments for further improvement of the paper:

(1) There are various types of mining activities related to the exploitation of natural resources. Underlying the activities are the spatial distribution of natural resources. For example, metals and minerals are of different distributions in China and as a result, the respective mining activities are in different places and of varying intensity. It seems that the paper does not present the big picture of spatial distributions of natural resources. Accordingly, the plot in Figure 1 is patchy, rather than comprehensive. Therefore, the authors may want to explicitly illustrate spatial distributions of natural resources as a big context of the literature survey work for the paper.

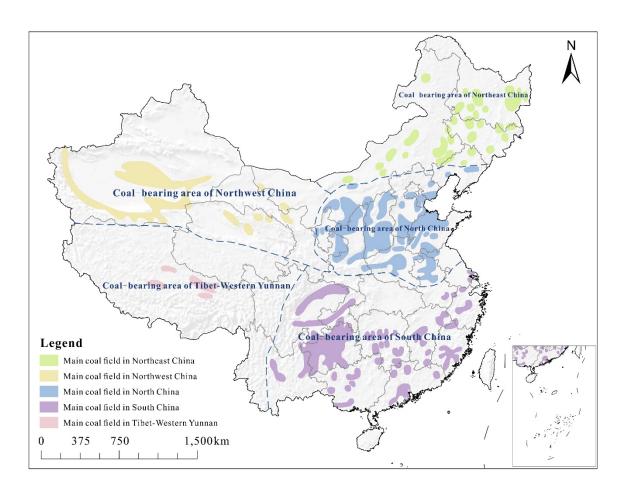
[Response] We appreciate your comment and have incorporated the suggestions into the revision. The belief information on natural resources in China has been presented in the revised manuscript (Lines 95-102) and in Section S1 of the Supplement (Lines 993-1032), which serves as the cornerstone for the database development, spatial pattern analysis, and risk assessment in the study. Specifically, Figs. S1 and S2 illustrate the spatial distribution and total sulfur content of coalbearing areas in China, and Fig. S3 exhibits the spatial distribution of the major non-ferrous mineral resources in China.

"China, the second-largest economy worldwide, has various and extensive mineral resources (Li et al., 2014). It has been demonstrated that there are 171 types of mineral resources in China, with proven reserves accounting for 12% of the world's mineral resources (Hu et al., 2009). Furthermore, China is one of the largest global producers and consumers of metals and metalloids, such as Fe, Mn, Zn, Pb, Sb, and Sn (Gunson and Jian, 2001). China's coal reserves of 143,197 million tons (Mt) rank fourth globally, while its annual production of 2,971 Mt leads worldwide (Blowes et al., 2014; Ai et al., 2023)."

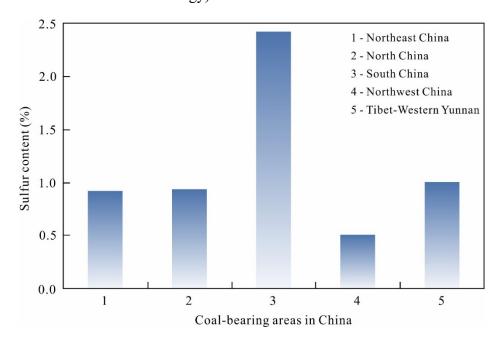
"The spatial distribution of coal fields shows significant regional differences, with dense concentrations in the coal-bearing areas of North and South China (Fig. S1). Among them, the southern Inner Mongolia, Shaanxi, Shanxi, and Henan provinces have the highest density of coal mines and mine production capacity. Besides, the coal resources of the junction of Anhui and Shandong provinces as well as Yunnan, Guizhou, Sichuan, and other provinces in southwest China are relatively rich (Xiao et al., 2021)."

"As shown in Fig. S3, China is rich in non-ferrous metal mineral resources. The predominant types are copper, lead-zinc, tin deposits, etc., mainly distributed in provinces such as Jiangxi, Yunnan, and Inner Mongolia. For example, the Dexing copper mine in Jiangxi province ranks as one of the largest copper deposits in China, while the Gejiu tin mine in Yunnan province is a world-renowned tin-producing area. Additionally, substantial precious metal mineral resources (gold and silver deposits) are predominantly located in Shandong, Henan, and Guizhou provinces. For example, the Zhaoyuan gold mine in Shandong province is a historically significant gold-producing region."

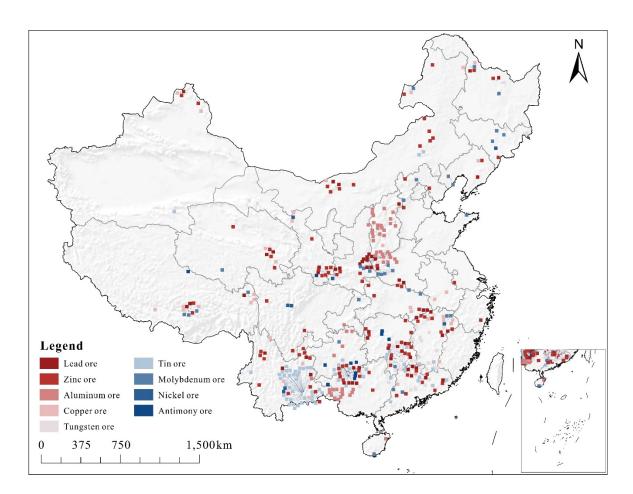
"It is noteworthy that the national mineral deposit database of China developed by Li et al. (2019), covering 232 mineral resources in 27,569 deposits in 29 provinces (cities or districts), is of great importance to study the national natural resources. It can help readers catch more authoritative information, such as ore species, deposit name, location, latitude (N), longitude (E), genetic type of deposit, paragenetic mineral, associated mineral, deposit scale, ore-forming age, and mining status, enabling comprehensive analysis of China's natural resources."



**Figure S1.** The spatial distribution of main coal-bearing areas in China (originated from China National Administration of Coal Geology).



**Figure S2.** The total sulfur content in different coal-bearing areas in China (adapted from Tang et al., 2015).



**Figure S3.** The spatial distribution of the major non-ferrous mineral resources in China (adapted from China Natural Resources Atlas, China Geological Survey, 2015, https://www.cgs.gov.cn/xwl/dzzl/201603/t20160309 304269.html).

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- (2) It is known that the mining of coal has a considerable impact on water quality, especially in West China. On the other hand, there is a lack of in-depth investigation of coal mining. Given the importance of coal mining and the existence of extensive studies, the authors may want to present a detailed analysis.

## References:

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[Response] Thank you for your constructive suggestions. An in-depth analysis of the considerable impacts of coal mining activities on water quality has been added to the revised manuscript based on the results obtained from the following references (Lines 100-105, Lines 551-555, and Lines 559-564):

"Coal is the predominant energy source for both domestic and industrial use in China, with reserves of 143,197 million tons (Mt) ranking fourth globally, while its annual production of 2,971 Mt leads worldwide (Blowes et al., 2014; Ai et al., 2023). However, coal extraction inevitably generates substantial amounts of mine water, resulting in a series of water environmental issues (Zhang et al., 2016; Qu et al., 2023). Current estimate shows a 2:1 mine water to coal production ratio, with approximately 2 tons of mine water produced per ton of extracted coal in China (Gu et al., 2021). Coal mining activities, especially those involving sulfide-rich coal mining, are intrinsically associated with acid mine drainage (AMD). Our investigation in this study focuses on mining regions in northern and southwestern China, which account for approximately 70% of the

national coal production. The spatial hotspots of mining-affected water pollution are mainly distributed in the southern regions, especially in Guizhou, Guangdong, Fujian, Jiangxi, Hunan, and Guangxi provinces/autonomous regions. 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). The main sulfide minerals in mine wastes are pyrite (FeS<sub>2</sub>) and pyrrhotite (Fe<sub>1-x</sub>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). Consequently, mining-affected water is characterized by the presence of diverse contaminants, including excessive sulfate, fluoride, and toxic heavy metals (Sun et al., 2020; 2025)."

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(3) There are serious concerns on heavy metal pollution in recent years. Previously, there have been a few review papers. What new insights (findings) does this paper make?

## References:

- Cheng, S., 2003. Heavy metal pollution in China: origin, pattern and control. Environmental science and pollution research, 10, pp.192-198.
- He, B., Yun, Z., Shi, J. and Jiang, G., 2013. Research progress of heavy metal pollution in China: Sources, analytical methods, status, and toxicity. Chinese Science Bulletin, 58, pp.134-140.
- Hu, H., Jin, Q. and Kavan, P., 2014. A study of heavy metal pollution in China: Current status, pollution-control policies and countermeasures. Sustainability, 6(9), pp.5820-5838.

[Response] The point is well taken. Based on your suggestions, we have reviewed the following references and cited them in the revised text and reference lists. Indeed, more than two decades ago, Cheng (2003) reviewed the sources of heavy metal pollution (*i.e.*, industrial emissions, wastewater irrigation, and waste fertilization) in the case sites, analyzed heavy metals in food and their transfer through the food web, and provided internationally available pollution control measures. About 10 years later, He et al. (2013) reviewed the status, sources, toxicity and potential risks, and possible reduction strategies of heavy metal pollution (especially for Pb, Hg, Cd, Cr, and As) in China. Also, Hu et al. (2014) reviewed the sources of heavy metal pollution (*i.e.*, waste gas, wastewater, and solid waste), discussed the policies (*e.g.*, the 12th Five-Year Plan on Prevention and Control of Heavy Metal Pollution) and challenges of controlling heavy metal pollution, and proposed corresponding countermeasures to mitigate heavy metal pollution by increasing the green GDP, reducing the heavy metals in fuel, using more renewable energy, and adopting market-based approaches.

Undoubtedly, the previous studies mentioned above have provided a solid basis for exploring the issues of heavy metal pollution originating from multiple pollution sources. In comparison to the previous studies, our study focuses on mapping mining-affected water pollution in China, elaborating on its status, patterns, risks, and implications. The new and unique contributions of the current study are elaborated on **Lines 744-772**:

"(i) establishing a national-scale high-quality database covering 8,433 surface water or groundwater samples (6,175 coal mine water samples and 2,258 metal mine water samples) from 298 mines (211 coal mines and 87 metal mines) in 26 provinces/autonomous regions of China; and (ii) filling the gap of the nationwide spatial patterns of water pollution and associated health risks from both coal and metal mining activities for the first attempt. Specifically, eight heavy metals (*i.e.*, Cr, Ni, Cu, Zn, As, Cd, Hg, and Pb) are considered in the current study based on a national-scale high-quality hydrochemical database. The new results show that Zn, Ni, and Cu are the

predominant contaminations contaminants of both coal and metal mines in China. The detectable concentrations of several heavy metals are higher in most metal mines than in coal mines, especially in mining-affected water with low pH (< 6.5). The order of detectable median values of water affected by coal mining is Zn (0.4211) > Ni (0.1796) > Cu (0.0431) > Cr (0.0080) > Cd (0.0036) >As (0.0034) > Pb (0.0023) > Hg (0.0004), while that of water affected by metal mining is Zn (7.200) > Cu (1.7325) > Ni (0.2142) > Pb (0.1498) > Cr (0.0500) > Cd (0.0383) > As (0.0281) >Hg (0.0090). In terms of spatial patterns, the pollution hotspots and potential risks of miningaffected water (with low pH, high sulfate, Fe, Mn, and heavy metals) are pronounced in the southern regions, especially in Guizhou, Guangdong, Fujian, Jiangxi, Hunan, and Guangxi provinces/autonomous regions. These phenomena are closely linked to the underlying mechanisms, such as climatic conditions, geological factors, and mining practices. Accordingly, the findings of the study yield critical insights for designing differentiated management measures and formulating spatially-adaptive pollution control strategies across three key dimensions, including geographic scales (site-specific scale, provincial scale, or national scale), mine types (coal or metal), and mining status (active or abandoned). This multidimensional framework enables policymakers to strategically balance the trade-off between green mining activities and human health priorities."

#### **Cited References:**

- Cheng, S.: Heavy metal pollution in China: origin, pattern and control. Environ. Sci. Pollut. Res., 10(3), 192-198. https://doi.org/10.1065/espr2002.11.141.1, 2003.
- He, B., Yun, Z.J., Shi, J.B., and Jiang, G.B.: Research progress of heavy metal pollution in China: Sources, analytical methods, status, and toxicity. Chin. Sci. Bull., 58(2), 134-140, https://doi.org/10.1007/s11434-012-5541-0, 2013.
- Hu, H., Jin, Q., and Kavan, P.: A study of heavy metal pollution in China: Current status, pollution-control policies and countermeasures. Sustainability, 6, 5820-5838, https://doi.org/10.3390/su6095820, 2014.

# Response to Referee #2's Comments

The manuscript aims to provide a comprehensive assessment of mining-affected water pollution across China by compiling a large dataset (8433 water samples from 298 mines). The study evaluates spatial patterns, assesses both carcinogenic and non-carcinogenic risks to human health, and discusses management implications for both coal and metal mining areas. While the work is well supported by extensive data and robust methodologies, questions remain regarding the novelty of the contribution, as the manuscript does not clearly delineate how its findings significantly extend beyond previous studies.

[Response] We sincerely thank you for your constructive and conscientious suggestions. Hereby we have fully incorporated and addressed all the comments in the revised manuscript and given a point-by-point response as below. In particular, a more explicit statement of our novel contributions relative to the existing literature (e.g., Cheng, 2003; He et al., 2013; Hu et al., 2013; Feng et al., 2014; Sun et al., 2025) has been added to the Conclusion (Lines 743-772) to address your concern that "the manuscript does not clearly delineate how its findings significantly extend beyond previous studies".

"It is noteworthy that previous studies predominantly concentrated on localized water pollution from individual coal or metal mines, while national-scale assessments have primarily addressed impacts exclusively attributed to coal mining activities. The new and unique contributions of the current study are: (i) establishing a national-scale high-quality database covering 8,433 surface water or groundwater samples (6,175 coal mine water samples and 2,258 metal mine water samples) from 298 mines (211 coal mines and 87 metal mines) in 26 provinces/autonomous regions of China; and (ii) filling the gap of the nationwide spatial patterns of water pollution and associated health risks from both coal and metal mining activities for the first attempt. Specifically, eight heavy metals (i.e., Cr, Ni, Cu, Zn, As, Cd, Hg, and Pb) are considered in the current study based on a national-scale high-quality hydrochemical database. The new results show that Zn, Ni, and Cu are the predominant contaminations contaminants of both coal and metal mines in China. The detectable concentrations of several heavy metals are higher in most metal mines than in coal mines, especially in mining-affected water with low pH (< 6.5). The order of detectable median values of water affected by coal mining is Zn (0.4211) > Ni (0.1796) > Cu(0.0431) > Cr(0.0080) > Cd(0.0036) > As(0.0034) > Pb(0.0023) > Hg(0.0004), while that of water affected by metal mining is Zn (7.200) > Cu (1.7325) > Ni (0.2142) > Pb (0.1498) > Cr (0.0500) > Cd (0.0383) > As (0.0281) > Hg (0.0090). In terms of spatial patterns, the pollution hotspots and potential risks of mining-affected water (with low pH, high sulfate, Fe, Mn, and heavy metals) are pronounced in the southern regions, especially in Guizhou, Guangdong, Fujian, Jiangxi, Hunan, and Guangxi provinces/autonomous regions. These phenomena are closely linked to the underlying mechanisms, such as climatic conditions, geological factors, and mining practices. Accordingly, the findings of the study yield critical insights for designing differentiated management measures and formulating spatially-adaptive pollution control strategies across three key dimensions, including geographic scales (site-specific scale, provincial scale, or national scale), mine types (coal or metal), and mining status (active or abandoned). This multidimensional framework enables policymakers to strategically balance the trade-off between green mining activities and human health priorities."

#### **Cited References:**

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- 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.

## **Novelty and Original Contribution**

Strengths: The study assembles a large national dataset and applies risk assessment models to evaluate health impacts, which is commendable. The spatial mapping of contamination hotspots and risk distribution provides valuable insights for policy-making.

[Response] No change needed. Thank you for your positive comments.

Concerns: One key issue is that the manuscript does not sufficiently highlight what is new compared to earlier studies. Although the scale of the data collection is impressive, the paper lacks a clear statement of its novel contributions relative to existing literature. The authors could enhance the manuscript by emphasizing unique aspects—such as new methodological approaches, previously unreported spatial trends, or innovative risk assessment strategies—that set this work apart.

[Response] The point is well taken. We have incorporated your concerns into the revision and a more explicit statement has been added to the revised Conclusion, highlighting the novelty and practical implications of our manuscript relative to the existing literature (Lines 743-772). See also the response to your general comment above.

## Methodological Rigor and Data Quality

Strengths: The methodology is generally robust, with clear criteria for data quality control and appropriate use of standard risk assessment models (e.g., those provided by the US EPA). The division of water samples (acidic vs. neutral/alkaline) and the differentiation between coal and metal mines are well executed.

[Response] No change needed. Thank you for your positive comments.

Suggestions: To further strengthen the paper, the authors should elaborate on how potential biases (e.g., variations in sample density among regions) were addressed. Additionally, more detailed statistical tests comparing water quality parameters between different mining types (such as using non-parametric tests) could provide further evidence for the observed differences.

[Response] Comment accepted. Indeed, there are potential biases caused by variations in sample density among regions. Therefore, our future in-depth research will attempt to address the biases by (i) combining the data mining and field sampling methods to investigate the potential contamination levels in more coal and metal mines across China; (ii) balancing the sampling density within each zone using bias correction techniques (*e.g.*, kernel density estimation and stratified spatial resampling) to ensure the data representation; and (iii) incorporating spatial uncertainty into the criteria to improve the spatial robustness for the assessments of mining-affected water pollution (Lines 710-716). Furthermore, we have added the results of non-parametric tests (*i.e.*, Mann-Whitney U-test and Spearman's rank correlation) to further support the differences observed in our study based on your suggestions (Lines 1086-1100):

"Non-parametric tests do not rely on assumptions about the distribution of the data and are suitable for non-normally distributed datasets or those containing outliers (Cardew, 2003). These methods statistically compare central tendencies, typically represented by medians, rather than means. The result of the Mann-Whitney U-test (p < 0.05) shows a statistically significant difference in the critical parameters (except Fe) of mining-affected water based on the different mine types (coal mine vs. metal mine), indicating the differences caused by geological factors, mining practices, surrounding environment, etc. Besides, Fig. S7 shows the Spearman correlation coefficients between the hydrochemical compositions in the mining-affected water. It can be seen that strong negative correlations are observed between pH and  $SO_4^{2-}$ , Fe, Mn, Al, and heavy metals

while positive correlations are observed between SO<sub>4</sub><sup>2-</sup> and metal components, implying that the spatial consistency of acid water, high sulfate, high Fe and Mn, and high heavy metal mining-affected water."

#### **Cited Reference:**

Cardew, P.T.: A method for assessing the effect of water quality changes on plumbosolvency using random daytime sampling. Water Res., 37(12), 2821-2832, https://doi.org/10.1016/S0043-1354(03)00120-9, 2003.

## **Presentation and Interpretation of Results**

Strengths: The results are logically presented, starting from the basic water quality parameters, moving on to spatial distribution patterns, and culminating in detailed risk assessments for different populations. Figures (e.g., maps and boxplots) support the textual description and help visualize the trends effectively.

[Response] No change needed. Thank you for your positive comments.

Suggestions: Although the numerical details are extensive, the manuscript may benefit from a more concise presentation. For example, summarizing key quantitative findings in a table could improve clarity. Additionally, while the spatial patterns are well described, a deeper discussion on the underlying geochemical or environmental processes that cause these trends would better contextualize the results.

[Response] Thank you for your insightful comments. To improve the clarity of the manuscript, key quantitative results (e.g., statistics of critical parameters for acid and neutral/alkaline water across different mines) have been summarized in Table 1 in the revised manuscript and Table S4 in the Supplement. Moreover, a further elaboration of the underlying mechanisms (e.g., geochemical conditions and environmental processes) driving the spatial patterns of mining-affected water pollution in China, especially in the highly polluted southern regions, has been added in Section 4.1 of the revised manuscript (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 wateroxygen 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 intrusionregression 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<sub>2</sub>) and pyrrhotite (Fe<sub>1-x</sub>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<sub>2</sub> by atmospheric oxygen can be expressed by Eqs. (8) - (11). 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)."

$$2FeS_2 + 7O_2 + 2H_2O \rightarrow 2Fe^{2+} + 4SO_4^{2-} + 4H^+$$
 (8)

$$4Fe^{2+} + O_2 + 4H^+ \rightarrow 4Fe^{3+} + 2H_2O \tag{9}$$

$$Fe^{3+} + 3H_2O \rightarrow Fe(OH)_3 + 3H^+$$
 (10)

$$FeS_2 + 14Fe^{3+} + 8H_2O \rightarrow 15Fe^{2+} + 2SO_4^{2-} + 16H^+$$
 (11)

Table 1. The statistics of mining-affected water in China (Units are mg/L except pH).

_		Acid mini	ng-affected wa	ater	Non-acid mining-affected water				
Item	Min	Median	Ave	Max	Min	Median	Ave	Max	
рН	1.20	3.52	4.04	6.50	6.51	7.80	7.85	12.60	
Na <sup>+</sup>	0.00	15.60	55.39	1613.00	0.23	156.01	403.20	9371.00	
$K^+$	0.00	3.30	6.41	172.00	0.00	3.11	7.10	419.00	
$Ca^{2+}$	0.83	284.80	284.33	987.97	0.00	64.10	119.76	4841.70	
$Mg^{2+}$	0.01	75.33	388.36	10992.00	0.00	19.01	52.85	12752.00	
Cl	0.00	3. 50	35.50	3097.40	0.00	51.68	314.21	26265.00	
SO <sub>4</sub> <sup>2-</sup>	0.56	1580.16	4648.54	181000.00	0.01	181.27	621.24	52915.00	
HCO <sub>3</sub>	0.00	1.89	48.65	769.00	0.00	253.23	343.13	4976.61	
$NO_3$	0.00	0.80	16.46	735.60	0.00	3.78	16.02	1774.95	
F	0.00	0.69	4.17	238.34	0.00	0.81	1.91	100.00	
Fe	0.0020	103.3000	520.4396	65250.0000	0.0000	0.1700	3.9788	495.4300	
Mn	0.0050	8.1080	71.1849	5050.0000	0.0000	0.2164	1258.9853	200000.0000	
Al	0.0077	53.9000	304.9210	13679.0000	0.0001	0.0350	0.5306	25.0000	
Cr	0.0000	0.0200	0.7725	52.2700	0.0001	0.0031	0.0081	0.3100	
Ni	0.0001	0.2166	1.6396	216.0000	0.0001	0.0050	0.0159	0.2260	
Cu	0.0002	0.8010	85.6400	9777.7700	0.0001	0.0100	0.0678	2.5600	
Zn	0.0026	2.3685	46.6335	1834.0000	0.0007	0.0391	0.5292	39.3000	
As	0.0000	0.0108	6.4969	641.7000	0.0001	0.0034	0.1362	13.0000	
Cd	0.0000	0.0220	0.6552	110.0000	0.0000	0.0004	0.0108	0.6677	
Hg	0.0000	0.0038	0.6997	13.3600	0.0000	0.0003	0.0196	0.7833	
Pb	0.0000	0.0700	0.5090	35.6800	0.0000	0.0012	0.0307	1.2200	

**Table S4.** The statistics of mining-affected water in China (Units are mg/L except pH).

	Acid mining-affected water						Non-acid mining-affected water					
Item	Coal mine			Metal mine		Coal mine			Metal mine			
	Min	Median	Max	Min	Median	Max	Min	Median	Max	Min	Median	Max
рН	1.90	4.50	6.50	1.20	3.10	6.50	6.51	7.82	11.51	6.51	7.70	12.60
Na <sup>+</sup>	0.02	18.55	1305.33	0.00	13.72	1613.00	0.23	234.75	7594.32	0.55	27.30	9371.00
$K^{+}$	0.04	3.50	37.00	0.00	2.99	172.00	0.00	2.84	164.42	0.20	3.80	419.00
$Ca^{2+}$	0.83	277.84	987.97	1.70	310.00	893.00	0.00	61.38	689.10	0.01	80.20	4841.70
$Mg^{2+}$	0.01	59.60	1665.00	0.10	89.52	10992.00	0.00	19.09	485.44	0.10	18.54	12752.00
Cl	0.06	2.51	477.24	0.00	9.20	3097.40	0.00	65.20	6462.75	0.35	19.00	26265.00
SO <sub>4</sub> <sup>2-</sup>	15.00	1381.59	17870.00	0.56	2982.00	181000.00	0.01	193.51	10110.00	0.09	157.41	52915.00
HCO <sub>3</sub>	0.00	0.00	532.96	0.00	15.51	769.00	0.00	280.60	4976.61	0.62	169.50	2482.00
$NO_3$	0.00	0.55	143.65	0.00	1.45	735.60	0.00	3.00	356.97	0.00	11.00	1774.95
F <sup>-</sup>	0.00	0.67	238.34	0.01	0.80	67.40	0.00	0.91	11.65	0.01	0.72	100.00

		Acid mining-affected water					Non-acid mining-affected water					
Item	Coal mine			Metal mine		Coal mine			Metal mine			
	Min	Median	Max	Min	Median	Max	Min	Median	Max	Min	Median	Max
Fe	0.0000	77.4100	2331.8560	0.0000	113.7700	65250.0000	0.0000	0.2500	205.0000	0.0000	0.0320	495.4300
Mn	0.0000	3.5000	88.4000	0.0000	15.8200	5050.0000	0.0000	0.0204	5.0200	0.0000	0.7612	200000.0000
Al	0.0000	12.8700	440.0000	0.0000	152.0000	13679.0000	0.0000	0.0200	25.0000	0.0000	0.0500	2.0900
Cr	0.0000	0.0080	0.1900	0.0000	0.0500	52.2700	0.0000	0.0022	0.3100	0.0000	0.0041	0.0909
Ni	0.0007	0.1796	2.7290	0.0000	0.2142	216.0000	0.0001	0.0040	0.2260	0.0000	0.0059	0.1200
Cu	0.0000	0.0431	18.5000	0.0000	1.7325	9777.7700	0.0001	0.0010	2.5600	0.0000	0.0180	1.1390
Zn	0.0026	0.4211	23.0000	0.0000	7.2000	1834.0000	0.0000	0.0048	0.9840	0.0000	0.0617	39.3000
As	0.0000	0.0034	2.1590	0.0000	0.0281	641.7000	0.0001	0.0016	0.3660	0.0001	0.0040	13.0000
Cd	0.0000	0.0036	0.1945	0.0000	0.0383	110.0000	0.0000	0.0000	0.0253	0.0000	0.0010	0.6677
Hg	0.0000	0.0004	0.0051	0.0000	0.0090	13.3600	0.0000	0.0001	0.7833	0.0000	0.0003	0.0050
Pb	0.0000	0.0023	7.4300	0.0000	0.1498	35.6800	0.0000	0.0003	1.2200	0.0000	0.0064	0.9400

## **Structure and Coherence of the Argument**

Strengths: The manuscript follows a conventional structure (introduction, methodology, results, discussion, conclusion) that makes it easy to follow. The discussion ties the findings back to the broader context of water pollution management.

[Response] No change needed. Thank you for your positive comments.

Suggestions: The transition between sections—especially from the results to the discussion—could be smoother. Explicitly linking how each result addresses the stated objectives would reinforce the coherence of the argument. Also, highlighting the novelty and practical implications of the work in the conclusion would help reinforce the manuscript's contribution.

[Response] Comment accepted. We have rewritten the transition between sections to reinforce the coherence of the argument. Moreover, a detailed discussion of the novelty and practical implications of our manuscript has been supplemented in the Conclusion to highlight the unique contribution and valuable addition to the field of environmental hydrology (Lines 743-772).

## Figures, Tables, and Visual Aids

Strengths: Visual aids are generally clear and provide a good overview of the data distribution and risk maps. The integration of detailed figures (such as spatial distribution maps and risk assessment graphs) adds significant value to the manuscript.

[Response] No change needed. Thank you for your positive comments.

Suggestions: Ensure that all figures have clear legends and consistent formatting. It might be beneficial to include a summary table that aggregates the key findings (e.g., median values of critical parameters across different mine types) to enhance readability.

[Response] Thank you for your constructive suggestions. We have reviewed/revised all figures to ensure that they have clear legends and consistent formatting. To improve overall readability, the summary tables (*i.e.*, Table 1 in the revised manuscript and Table S4 in the Supplement) showing the statistics of critical parameters for different mine types have been added to the current revision.

## Language and Style

Strengths: The manuscript is written in clear, professional English with an appropriate academic tone. Technical terms are defined upon first use, and the text is generally free of major grammatical errors.

[Response] No change needed. Thank you for your positive comments.

Suggestions: A few sentences could be simplified to improve readability. In particular, some complex sentences in the introduction and discussion might be broken into shorter, more digestible statements. Maintaining consistency in terminology (for instance, ensuring that terms like "differentiated management" are clearly defined) will also help in reinforcing the manuscript's clarity.

[Response] Comment accepted. We have simplified some complex sentences in the Introduction and Discussion to improve the readability of the manuscript. Moreover, a clear definition of terms like "differentiated management" has been added in the revised manuscript to reinforce the manuscript's clarity (Lines 650-655):

"The differentiated management mentioned in the current study is an optimized regulatory paradigm that customizes strategies to mine types (coal vs. metal) and operational status (active vs. abandoned) based on hydrogeological conditions, pollution source characteristics, and multisystem sustainability requirements. The initiative aims to implement targeted intervention and precise prevention/control to mitigate pollution risks, restore and enhance ecological functions, while concurrently safeguarding human health."

#### Conclusion

The manuscript presents an extensive dataset and a rigorous analysis of mining-affected water pollution in China, offering useful insights for environmental management and policy-making. However, the work would benefit from a more explicit discussion of its novelty compared to previous studies. Clarifying and emphasizing the unique contributions—whether in data scale, methodological advancements, or new insights into spatial and health risk patterns—would significantly strengthen the paper. With these revisions, the manuscript could represent a valuable addition to the field of environmental hydrology.

[Response] We sincerely appreciate your conscientious and constructive comments. A more explicit discussion of the novelty compared to previous studies has been added to the revised manuscript (Lines 743-772), to provide new insights into the spatial patterns and health risks of mining-affected water pollution at the national scale, and to clarify and emphasize the unique contributions of our study. We believe that your insightful comments on 'Novelty and Original Contribution', 'Methodological Rigor and Data Quality', 'Presentation and Interpretation of Results', 'Structure and Coherence of the Argument', 'Figures, Tables, and Visual Aids', and 'Language and Style' have led to significant improvements of the 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<sub>2</sub>) and pyrrhotite (Fe<sub>1-x</sub>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<sub>2</sub> by atmospheric oxygen can be expressed by Eqs. (8) - (11). 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)."

$$2FeS_2 + 7O_2 + 2H_2O \rightarrow 2Fe^{2+} + 4SO_4^{2-} + 4H^+$$
 (8)

$$4Fe^{2+} + O_2 + 4H^+ \rightarrow 4Fe^{3+} + 2H_2O$$
 (9)

$$Fe^{3+} + 3H_2O \rightarrow Fe(OH)_3 + 3H^+$$
 (10)

$$FeS_2 + 14Fe^{3+} + 8H_2O \rightarrow 15Fe^{2+} + 2SO_4^{2-} + 16H^+$$
 (11)

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- 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<sub>p</sub>), reference dose (RfD<sub>o</sub>), gastrointestinal digestion coefficient (ABS<sub>GI</sub>), 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 516-520):

Table S2. The main parameters used for human health risk assessment.

D	Diti	11. 4	Va	C	
Parameter	Description	Unit -	Adult	Children	Source
IR	Ingestion rate	L/d	2.50	0.78	[1], [2]
EF	Exposure frequency	d/yr	350	350	[1], [2]
ED	Exposure duration	yr	24	6	[2]
ET	Time of contact	h/d	0.58	1.00	[3], [4]
SA	Skin surface area	$cm^2$	19652	6365	[1], [2]
CF	Conversion factor	L/cm <sup>3</sup>	0.001	0.001	[2], [5]
BW	Body weight	kg	70	15	[1], [3], [4]
AT	Averaging time <sup>a</sup>	d	8760	2190	$ED \times 365 \text{ d/yr}$
A1	Averaging time b	d	25550	25550	70×365 d/yr

Note: <sup>a</sup> averaging time used for non-carcinogenic risks (NCRs), and <sup>b</sup> averaging time used for carcinogenic risks (CRs), which is equal to a lifetime (70 yr in the study) ×365 d/yr. The parameter values used in the study are obtained from the following literature sources: [1] Meng et al. (2024); [2] Shi et al. (2023); [3] Tong et al. (2021); [4] Wang et al. (2021); and [5] Yuan et al. (2023).

**Table S3.** The values of main parameters including permeability coefficient of skin  $(K_p)$ , reference dose  $(RfD_o)$ , gastrointestinal digestion coefficient  $(ABS_{GI})$ , and slope factor (SF) for each element.

Parameter	$K_p$	$RfD_o$	$ABS_{GI}$	SF	Source	
1 arameter	(cm/h)	(mg/kg·d)	(-)	(kg·d/mg)		
Fe	0.001	0.7	0.2	-	[1], [2], [3], [4], [6]	
Mn	0.001	0.024	0.04	-	[1], [2], [3], [4], [6]	
Cr	0.002	0.003	0.025	0.5	[1], [3], [6], [7]	
Ni	0.0002	0.02	0.04	-	[1], [2], [3], [4], [6], [7]	
Cu	0.001	0.04	0.2	-	[1], [2], [3], [4], [6], [7]	
Zn	0.0006	0.3	0.2	-	[1], [2], [3], [4], [5], [6]	
As	0.001	0.0003	1	1.5	[1], [3], [7]	
Cd	0.001	0.0005	0.05	0.38	[2], [3], [4], [6]	
Pb	0.0001	0.0014	0.3	-	[1], [3], [6]	

Note: The parameter values for each element are obtained from the following literature sources: [1] Meng et al. (2024); [2] Shi et al. (2023); [3] Tong et al. (2021); [4] USEPA (2002); [5] USEPA (2014); [6] Wang et al. (2021); and [7] Zheng et al. (2023).

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- 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.