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Spatial and temporal distribution of Mo in the overlying water of a reservoir downstream from mining area

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ABSTRACT

This study aimed to evaluate the spatial and temporal variations of molybdenum (Mo) in the downstream water body of a Mo mine during three hydrologic periods (wet, dry and medium seasons). The physical properties in Luhun Reservoir reflected seasonal variations in different hydrological periods. The redox potential (ORP) and dissolved oxygen (DO) increased in the dry season. The concomitant decrease in temperature (T), conductivity (COND) and total dissolved solids (TDS) were lowest in the wet season. The pH value did not change significantly during the three hydrologic periods. The distribution of Mo in the dry season was high in upstream and low in downstream areas, which was significantly different from that of the wet and medium seasons. The total Mo concentration in wet (150.1 µg/L) and medium season (148.2 µg/L) was higher than that in the dry season, but the TDS (288.3 mg/L) and the percentage dissolved Mo (81.3%) in overlying water was lowest in the wet season. There was no significant relationship between the dissolved Mo and the total Mo with TDS. In the dry season, the mean total Mo concentration was 116.3 µg/L, which was higher than the standard limit value (70 µg/L) for drinking water (US EPA-United States Environmental Protection Agency recommended value 40 µg/L). Non-point source pollution is the main characteristic of mining area pollution, which was closely related to rainfall. Thus, the Luhun Reservoir contains substantial Mo pollution, which was a significant concern given that it is used as a source of drinking and irrigation water.

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Introduction

Contamination with heavy metals (trace metals) and their bioaccumulation in the environment is detrimental to hu-

man and environmental health (Vicente-Martorell et al., 2008; Yousaf et al., 2016). The release of metal contaminants into water can significantly affect the ecology and subsequently lead to the degradation of water quality as well as the whole ecosystems (Acevedo-Figueroa et al., 2006; Zhang et al., 2009; Wang et al., 2015). Mining operations are vital to sustaining our industrial development, but unfortunately increases the risk of detrimental effects of heavy metal contamina-

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tion in areas such as lake reservoirs downstream of mining areas and stream waters in a mining activity contaminated site (Rodríguez et al., 2008; Gao et al., 2016; Li et al., 2016; Sharon et al., 2018). The aqueous phase provides a mobile medium for metal chemical reactions and metal circulation through three significant pathways: organometallic ion complexes, inorganic metal ions, and metal ions accumulated by phytoplankton; and the water salinity, water silicate, water Chlorophyll and metals (Cu, Pb, Zn, Cr, As, Fe, Mn, Ni, Co, Cd) are impacted differently during the dry, wet season and medium seasons (Campanha et al., 2012; Heijerick et al., 2012; Bebianno et al., 2015). Pollution may be influenced by changes in climate for mines in some regions, for example in wet season the heavy metal pollution of water in mining areas is highly susceptible to rainfall (Yang et al., 2020; Shenai-Tirodkar et al., 2018). Therefore, it is necessary to study the seasonal variation of heavy metals during different hydrological periods in reservoirs downstream of mining areas.

There have been many studies on heavy metals migration and transformation processes, including desorption, adsorption, precipitation and biological absorption in water bodies, sediments and organisms, especially for those metals that show high toxicity and long-term persistence within the food chain (Silva et al., 2009; Soma and Kumar, 2013). However, few studies have focused on Mo pollution affected by seasonal changes, although Mo compounds in drinking water are easily absorbed, and excessive intake of Mo can lead to gout or other symptoms (Silva et al., 2009; Diop et al., 2015; Zhang et al., 2019). Therefore, we selected the Luhun Reservoir as our study area, which is a reservoir downstream of the largest Mo mine in China, and collected samples to monitor the Mo and physical properties during three hydrological periods. This approach allowed us to examine the water quality from mining operations in Luhun Reservoir throughout different seasons (Ip et al., 2007; Han et al., 2017), by exploring the pollution characteristics of water in the reservoir. We evaluated positive protective measures for waste containment and management practices of mining sites in this region.

1. Material and Methods

1.1. Study area

Sampling sites were located in Luhun Reservoir, Henan Province, China as shown in Fig. 1, in the middle reaches of the Yi River in the Yellow River basin. The average depth of the Luhun Reservoir is 9.6 m, the maximum depth is 31.0 m; and the reservoir length is 12.5 km, width is 3.5 km. Luhun Reservoir is in a semi-arid and semi-humid area, which belongs to the temperate continental monsoon climate. The annual average temperature is about 14.1°C and the annual average rainfall is 791 mm. More than 60% of the annual rainfall is concentrated in the flood season from June to October.

The Luhun Reservoir basin and its surrounding areas (Songxian and Luanchuan counties) contain more than 40 Mo metal mineral deposits, such as the Yuchiling, Fantaigou, Angou and Luanchuan Mo mines. Other metallic ores (lead, zinc, gold and iron ores) have been identified in the region with more than 180 mineral producing areas. The geochemistry of

the deposit being mined and the chemical and ecological nature of the surrounding water bodies were associated with risks of affected water quality. This study was focused on overlying water of the Luhun Reservoir; basic information on the distribution of sampling sites is shown in Fig. 1.

There were obvious distribution characteristics of the dry, med and wet seasons (flood season) in Luhun Reservoir (Fig. 2). However, the peaks were unstable and mainly occurred from June to October each year, with high summer rainfall which belonged to the wet season. The maximum monthly intake was up to 590 million m³ and the minimum inflow was as low as 500,000 m³ per month, with extensive fluctuations observed. The dry season extended from November to February and was characterized by low intake; whereas the intermediate season extended from March to July. The change in water intake was a key factor affecting water quality, mainly involving non-point source pollution, dissolved or solid pollutants. Meanwhile, many mining, processing and smelting enterprises were distributed across the upper reaches of the Luhun Reservoir, which was characterized by serious non-point source pollution. The scouring effect of summer rainfall runoff mainly entered the upstream river water through low-lying areas, causing serious reduction in water quality.

1.2. Sample collection and analysis

Water samples and surface sediment samples were collected from each sampling site during the three hydrological periods: mid-August, 2018, mid-January, 2019 and mid-April, 2019 for wet season, dry season and medium seasons respectively. The overlying water samples of 500 mL were collected from the Luhun Reservoir, which contained a mixture of surface, middle and bottom overlying water samples. The water samples were collected and stored at 4°C for analysis. Conductivity (COND), total dissolved solids (TDS), redox potential (ORP), pH, and dissolved oxygen (DO) were analyzed in the field using a portable meter YSI-556 (YSI-556; YSI Inc. Yellow Springs, OH, USA).

The Milli-Q water were prepared (>18 MΩ cm, Merck Millipore, Billerica, MA). All sampling bottles were pre-cleaned with 10% (V/V) HNO₃ and rinsed with Milli-Q water. Sample aliquots were filtered through 0.45 μm membrane filters to determine the concentration of dissolved Mo. Samples for determination of total Mo concentration were microwave digested in an HCl-HNO₃-HF-HClO₄ mixture (MARS Xpress, CEM Corp., Matthews, NC). The Mo concentration was determined using an inductively coupled plasma mass spectrometer according to standard methods. Blanks and each set of samples were evaluated in duplicate and the relative deviation was < 5% (Rauret et al., 1999; Pueyo et al., 2008). The protocol included repetitive analyses of procedural blanks and a standard (GBW07427, National Land Resources Geophysical Research Institute) for quality control.

1.3. Statistical analysis

The sample sites and sampling were represented using Google Earth and ArcGIS 9.3.

The Origin Pro 8.5 software was used to plot the data (OriginLab Corp., Northampton, MS, USA). Data were analyzed

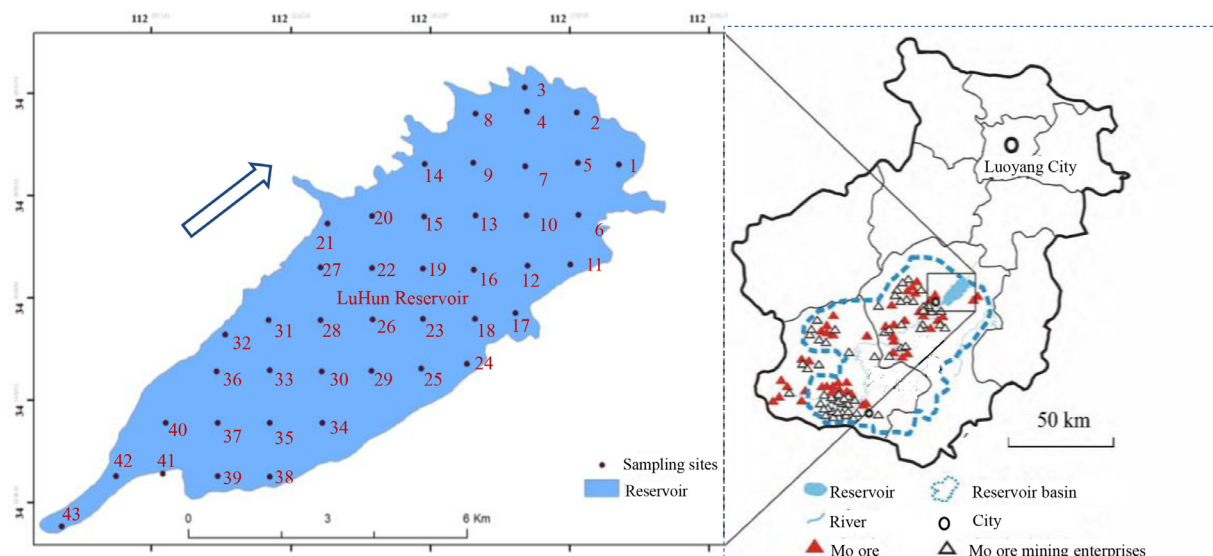


Fig. 1 – Locations of sampling sites, Mo mineral deposits and operating statuses of mines.

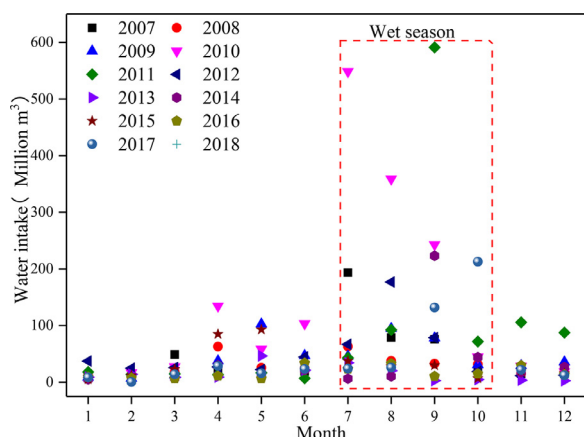


Fig. 2 – Annual variation characteristics of water intake in LuHun Reservoir from 2007 to 2018.

using SPSS 16.0 and two-sample t-tests ($p < 0.05$) were used to determine differences between mean values (IBM Corp., Armonk, NY, USA).

2. Results and Discussion

2.1. Physical properties of overlying water

The physical properties of overlying water samples are shown in Fig. 3. The mean pH values of the water in LuHun Reservoir were 8.3, 8.2 and 8.4 for wet season, dry season and medium season respectively. The range of variation pH values of water in medium season was relatively small (± 0.13). The water temperature mean value in LuHun reservoir in wet (summer) season was 29.8°C. In dry (winter) season, the mean water temperature ranged from 4.3 to 5.3°C, and the mean temperature was 4.8°C. In medium season, the water temperature mean

value was 14.4°C. The CODN of the water body was the highest in medium (502.3 $\mu\text{S}/\text{cm}$) season and the lowest in wet (435.5 $\mu\text{S}/\text{cm}$) season. The TDS was 288.3, 333.6 and 346.3 mg/L in wet season, dry season and medium seasons, respectively. The reservoir water TDS was lower than that required by the sanitary standard for drinking water (Yang et al., 2020). The mean ORP value was 160.1 and 240.2 mV in wet season and dry season, respectively. The DO ranged from 6.5–11.2 mg/L from wet season to medium seasons, almost reaching the value for surface water class III water standard (5 mg/L), indicating reasonable water quality in the reservoir (Lin et al., 2020).

2.2. Total Mo concentration in overlying water

The total Mo concentration in the water body of LuHun Reservoir was relatively high in the wet season with an average value of 150.1 $\mu\text{g}/\text{L}$ (Fig. 4a); the total concentration was higher than the standard limit for Mo in surface water (70 $\mu\text{g}/\text{L}$) and the United States Environmental Protection Agency recommended value of 40 $\mu\text{g}/\text{L}$ (Soma and Kumar, 2013; Pichler et al., 2016). Upstream Mo mines and mining enterprises in the reservoir area contributed to the Mo in the water body. The total amount of heavy metals released from tailings was related to the climate, except that temperature and humidity were the main factors affecting the oxidation of sulfide minerals, and the periodicity of precipitation season was also an important condition that affected the heavy metal release from tailings in mining areas. The summer rainfall was relatively large, which resulted in non-point source pollution with rain-water confluence into the water body and eventually into the reservoir area (Varol, 2013). According to the spatial distribution, the total Mo concentration in the upper part of the LuHun Reservoir was higher than that in the downstream part of the reservoir and inclined to decrease with the downstream flow direction.

In dry season (Fig. 4b), the total Mo concentration was 48.33–156.02 $\mu\text{g}/\text{L}$, with a mean value of 116.3 $\mu\text{g}/\text{L}$; this was higher than the standard limit for Mo in surface water

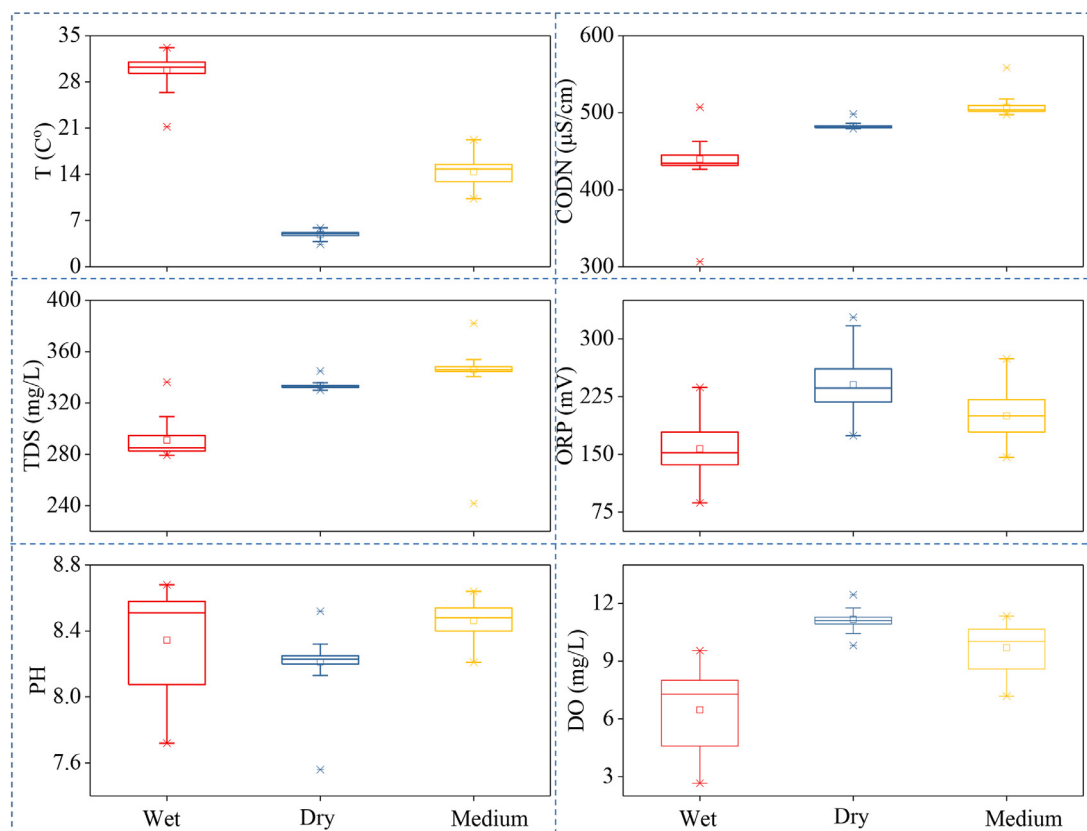


Fig. 3 – Physical properties of overlying water from Luhun Reservoir

(70 $\mu\text{g/L}$). Apart from the upper part of the reservoir, the total Mo concentrations of other sampling points in the dry season were higher than the standard limit value (70 $\mu\text{g/L}$). Based on the spatial distribution of total Mo concentration, the concentration in water tended to increase, with upstream concentrations significantly lower than those of the middle and downstream concentrations. The mean total Mo concentration in the medium season was 148.2 $\mu\text{g/L}$ (Fig. 4c), which was also higher than the standard limit of Mo for surface water (70 $\mu\text{g/L}$). The distribution characteristics of Mo in the medium season were similar to those of the wet season, i.e., high in the upstream and low in the downstream part of the reservoir. The mean total Mo concentrations in the water of Luhun Reservoir were similar in the wet and medium seasons (150.1 and 148.2 $\mu\text{g/L}$, respectively), both of which were higher than that in the dry season (116.3 $\mu\text{g/L}$).

The climate of Luanchuan was relatively dry, and precipitation mainly concentrated in June to October. Heavy metal sulphide oxidation occurred via neutralization reaction in sulphide minerals oxidized product because there was no water soluble dissolve for migration. During the wet season, the accumulation of soluble elements or ion released quickly and brought greater pressure on the downstream. The reason for the high concentration of Mo in the Luhun Reservoir from June to October was related to the high temperature and accelerated oxidation rate of bauxine, as well as the rapid leaching release of the accumulated Mo with coming of the rainy season. However, since the Mo mine has existed for an ex-

tensive period of time, previous mining has caused significant sedimentation of Mo in the riverbed. Consequently recontamination of sediments by storm water was a major concern when evaluating the potential effectiveness. There were studies showed that storm water contaminant mass and sediment recontamination differed from other sources, and different heavy metals exhibited different characteristics. For example, Cu was not easily released from the sediment, which in overlying water was mainly in particulate fraction; meanwhile Zn was mainly in dissolved form (Yuki et al., 2020; Tian et al., 2020; Drygiannaki et al., 2020). The interactions between the mining industry and pollution characteristics of water and sediment were often complex and site specific, and the relationship between molybdenum concentration in river and the mining area needed to be further studied.

2.3. Dissolved Mo concentration in overlying water

The dissolved Mo concentration was analyzed to assess the morphological characteristics of Mo in water and the influence of hydrologic changes (Fig. 5a). There was little difference in the mean percentage of dissolved Mo in the water of Luhun Reservoir in the dry and medium seasons (93.2% and 94.3%, respectively), while the percentage was substantially lower in the wet season (81.3%). This suggested there was substantial seasonal variation in river water quality in the Luhun Reservoir basin, with large amounts of Mo and non-point source pollutants including solid particles migrating to the down-

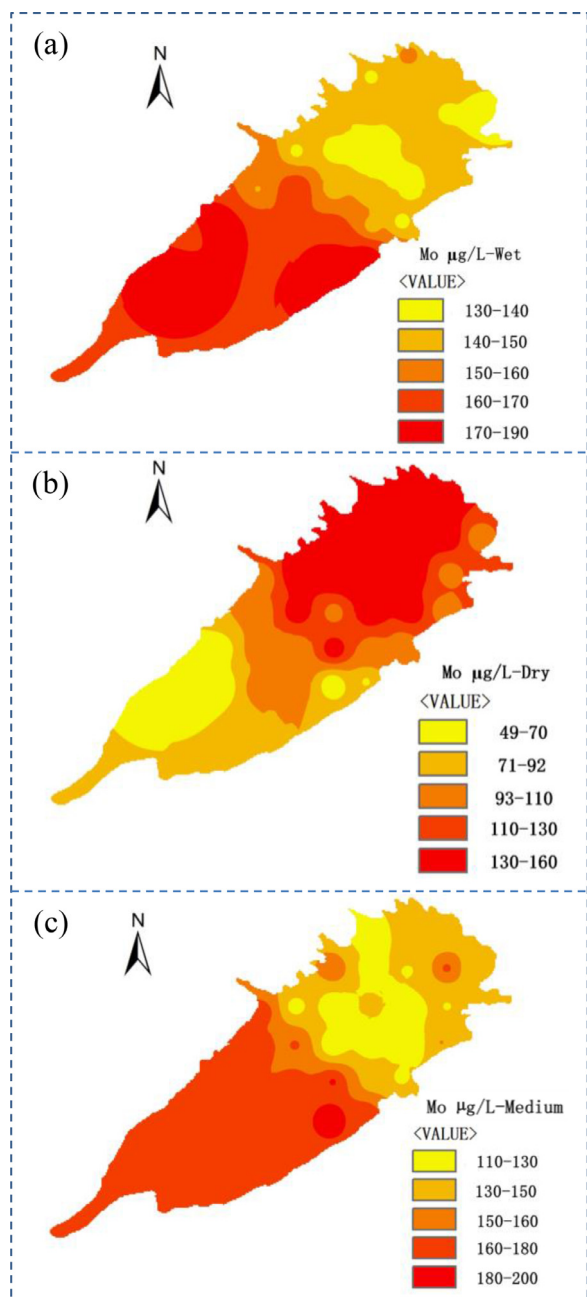


Fig. 4 – Distribution of Mo total concentration in overlying water of the Luhun Reservoir a: wet season; b: dry season; c: medium season

stream region; the water quality presented the characteristics of serious pollution during the wet season (Ma et al., 2015; Israel et al., 2018). In the dry and medium seasons, the water intake was small. Therefore, the percentage of dissolved Mo in water was higher in the dry and medium seasons than in the wet season.

The seasonal variations in the percentage of dissolved Mo were similar to the characteristics of TDS in overlying water (Fig. 6b). To determine whether these two characteristics were related, the distribution characteristics of TDS in overlying water were analyzed. TDS refers to the total amount of

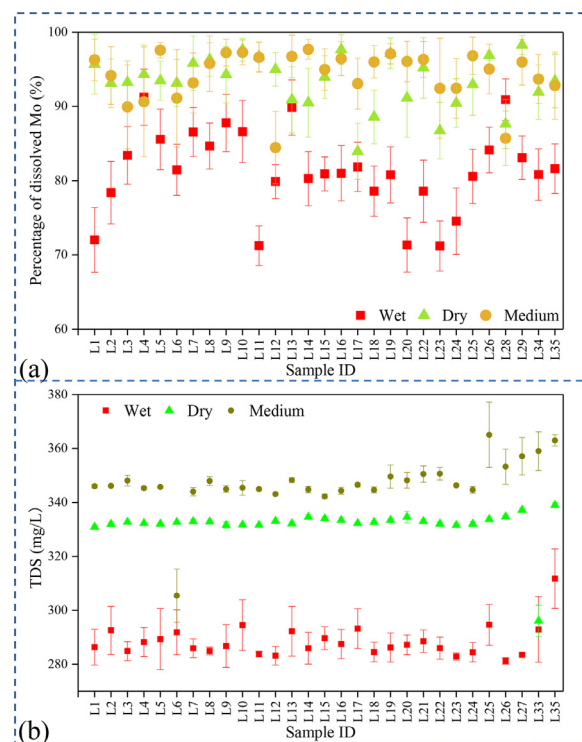


Fig. 5 – Distribution characteristics of percentage dissolved Mo and TDS in overlying water of the Luhun Reservoir

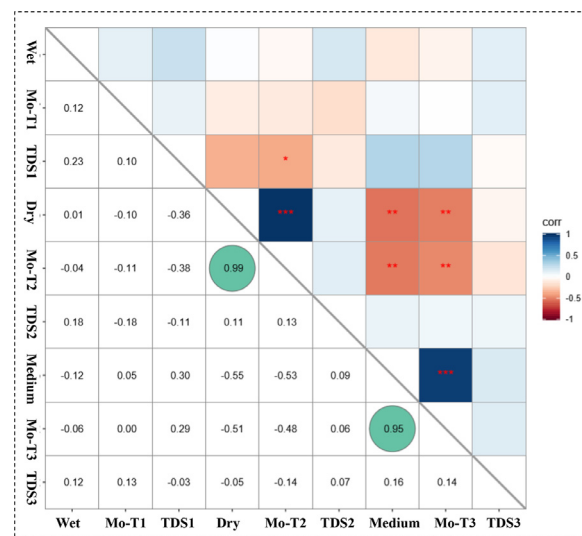


Fig. 6 – The relationships among seasonal mean values of total Mo concentration, dissolved Mo concentration and total dissolved solids (TDS) by Pearson correlation coefficients ($n = 57$).

solutes in water, including both inorganic and organic content (Drover et al., 2019; John and Chellappa, 2019). Conductivity was utilised to understand the salinity in the solution; generally, the higher the conductivity, the higher the salinity and the higher the TDS (Juan et al., 2019; Yang et al., 2020). The national standard (GB5749-2006) "sanitary standards for

drinking water" has limited requirements for TDS of drinking water to: $\text{TDS} \leq 1000 \text{ mg/L}$ (Gao et al., 2015).

The water body of Luhun Reservoir had the highest TDS in Med of 346.3 mg/L and the lowest TDS in wet of 288.3 mg/L; the most uniform distribution of TDS was observed in Dry with an average value of 333.6 mg/L. The TDS distribution in Luhun Reservoir was relatively uniform in wet season, with the maximum value of 314.5 mg/L, minimum value of 281.4 mg/L, and mean value of 287.4 mg/L. In dry season, the TDS of Luhun Reservoir was evenly distributed. In medium season, the range of TDS was 342.1–365.1 mg/L, which was very similar to the seasonal variations in percentage of dissolved Mo in overlying water. Generally the TDS in the reservoir area water met the requirements of the sanitary standard for drinking water.

2.4. Relationships between Mo and TDS in overlying water

The Pearson correlation coefficient was used to analyze the relationships among the seasonal total Mo concentration, dissolved Mo concentration and TDS in overlying water (Fig. 6). There were significant positive correlations between the Dry (dissolved Mo in dry season) and Mo-T2 (total Mo in dry season), and between the Medium (dissolved Mo in medium season) and Mo-T3 (total Mo in medium season), which were similar to the percentage dissolved Mo in overlying water. There was a significant negative relationship observed among Dry, Medium, Mo-T2 and Mo-T3, as shown in the distribution of the total Mo concentration in overlying water. However, there was no relationship observed among Wet (dissolved Mo in wet season) and Mo-T1 (total Mo in wet season) or TDS1, implying that there were more solid particles in wet season, which had a great influence on the concentration of dissolved Mo in water; in addition, the presence of other soluble salts in water, such as nitrogen, phosphorus and other ions, meant that TDS was not an effective indicator for the Mo concentration in water (Huang et al., 2012).

Moreover, adsorption mechanism was the important mechanism of heavy metal retention in tailings; many studies suggested that the adsorption mechanism in heavy metals pollution played an irreplaceable role. Acidic range promoted the adsorption of anionic, and alkaline range promoted that of the cation adsorption (Soma and Kumar, 2013; Pichler et al., 2016). Fe and Mn oxides (hydrated oxides) adsorbed Mo in a pH range different from Cu and Zn. In the oxidation environment with $\text{pH} < 8.0$, Fe and Mn colloids were the most powerful colloids for adsorption of Mo, especially at pH 3–5. At $\text{pH} > 8.0$, manganese colloid could hardly absorb ionic Mo. Mo on the colloid exhibited desorption back into aqueous phase at $\text{pH} > 7.0$. The mean pH values of the water in Luhun Reservoir were 8.3, which was the main reason of no relationship observed among Wet and Mo-T1 or TDS1.

3. Conclusions

This study analyzed the annual variation in water intake and seasonal variation in the total Mo concentration, percentage of dissolved Mo, and TDS in different seasons. The results showed that the Mo concentration was high in Wet and Med

seasons, the TDS was higher in dry and medium seasons, and the DO and ORP were higher in the dry season, the pH was relatively stable throughout the year. The variation in total Mo concentration, percentage dissolved Mo and TDS in overlying water changed significantly during three hydrologic periods. Because of the large amount of non-point source pollutants that migrated to the downstream in the wet season, including many solid particles, the percentage of dissolved Mo in water was lowest in the wet season. However, there were no significant correlations between the Mo concentration and TDS in water, so the TDS was not an effective indicator for the Mo concentration in water. The concentration of precipitation in Luanchuan mine was concentrated, and the high Mo concentration of heavy metal can be detected in the downstream of the mine in the rainy season. If measures were taken to prevent the release of heavy metal Mo leaching in the rainy season, it will play an excellent role in relieving the pollution pressure of Mo in the downstream environment.

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Appendix A Supplementary data

Supplementary material associated with this article can be found, in the online version, at [doi:10.1016/j.jes.2020.09.033](https://doi.org/10.1016/j.jes.2020.09.033).

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