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Summary of the Dissertation

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学位論文題名

The title of the doctoral dissertation

Sustainable management of a legacy mine considering long-term contaminant transport

（汚染物質の長期的な移行を考慮した休廃止鉱山の持続可能な管理）

1. Introduction

Acid mine drainage (AMD), also known as acid rock drainage (ARD), having characteristic of low pH and higher dissolved metals, is usually associated with anthropogenic mining activities and tunnel constructions, which often involve the contamination of both surface and groundwater resources (Asta et al., 2010; Candeias et al., 2014; Concas et al., 2006; Jennings et al., 2008; Stumm & Morgan, 1996). The drastic effects of mining activities on natural environments remain prominent, even after the mining operations have ceased for centuries or decades. Mining activities (excavation of ore deposits, mineral beneficiation, and disposal of mine wastes) are of major concern in the formation of AMD.The sulfide minerals, such as pyrite (FeS_2), chalcopyrite ($CuFeS_2$), galena (PbS), sphalerite (ZnS), are naturally formed ore minerals that tend to be stable under reductive environmental conditions. As economic point of views, the oxidation of sulfide minerals can be a good indicator of secondary sulfide ore enrichment in the context of hydrometallurgy in mine sites. On the other hand, for the natural environment, it becomes a source of soil and water contamination (Rimstidt et al., 1994). For example, when pyrite is in contact with water and oxygen, it can release oxidation products of ferrous ion (Fe^{2+}), sulfate ion (SO_4^{2-}), and hydrogen ion (H^+), leading to the further oxidation and hydrolysis reactions. The released ferrous (Fe^{2+}) can be further oxidized to ferric ion (Fe^{3+}) and proceed to hydrolysis or precipitate as Fe $(OH)_3$ but still liberating H^+ and continuously acidify the mine water. Without minimizing the contamination, these acidic leachates will migrate over several hundred meters and finally enter the ecological and biological systems. One of the AMD problems is that even after the mine closure, the remaining sulfide minerals continue to induce oxidation, drastically affecting the surrounding soil and water qualities, as observed in the Iron Mountain Mine and Leviathan Mine in USA (Druschel et al., 2004), the Iberian Pyrite Belt (IPB) in Spain (Espana et al., 2005), Panasqueira mine in Portugal (Candeias et al., 2014), and the Yatani mine and Shimokawa mine in Japan (Khoeurn et al., 2019; Tomiyama et al., 2019).

Numerous AMD investigations worldwide have demonstrated that even after the mine closure, reactive sulfide minerals continue to have the potential to produce AMD, which has a significant impact on the aquatic ecosystems and human health. Enormous novel approaches have also been proposed in AMD remediation strategies regarding chemical, physical, and biological methods by considering the paramount roles of treatment efficiency, economic aspects, and the critical insight into achieving a sustainable environment. In various aspects of hydrogeologic investigations, artificial and natural tracers are used worldwide to address contaminant hydrology because of their effective potential in solving anthropogenic pollution problems, groundwater dating, tracing the paleo-history of water resource, and their mixing effects have been evaluated (Cowie et al., 2014; Nigro & Barbieri, 2017; Silva & Cota, 2021; Solomon et al., 1995). Implementing effective AMD remediation strategies requires a proper understanding of the sources of AMD and its geochemical control governing the evolution of mine water quality, as well as its hydrological pathways facilitating the transport of contaminants to nearby water resources. These will provide valuable insights into long- term AMD management, including the reduction of the financial burden and selection of alternative sustainable remediation strategies. There are a few studies regarding the management of legacy mine sites for long-term evolution of AMD due to a complex hydrological mine site, with many possible sources of AMD; mine adit, tailing dam, and their control on the evolution of groundwater quality.

By taking this opportunity, one of the legacy mine sites in Japan is chosen (Fig. 1). In 1720, the mine was initiated as an Au mine, and the proceeding years followed the production of Pb and Cu with a refining process in 1844. There

are several shafts and drifts for over 300 years. Due to higher tonnage and grade of ores, it became one of the most important minerals with economic relevance in Japan (JOGMEC, 1997). However, in 1957, the mine site finally led to an end because of a decline in ore grade and tightening environmental regulations in Japan. During the mine operation, the mine tailings were discharged into the tailing dam and sedimentation impoundment located in the upper and the middle parts of the mine site. They were systematically designed with seepage drainage tunnels and vegetation cover along the slopes to protect the tailing liquefaction and landslide problems. According to historical monitoring data, after the mine closure, the mine water changed to pH ≈ 3.7 with high heavy metal concentrations including Zn (≈ 14 mg/L), Cu (≈ 4.2 mg/L), and Pb (≈ 0.6 mg/L) in 1981 (JOGMEC, 1997). Then, the government of the city started to monitor and treat the mine water by active treatment methods using slaked lime. This has been operated by the local government; however, due to the uncertainty of the treatment timeframe and the burden of the treatment costs over time, alternative effective methods need to be evaluated. Therefore, this study is to implement a long-term sustainable treatment method which is cost-effective and environmentally sustainable.

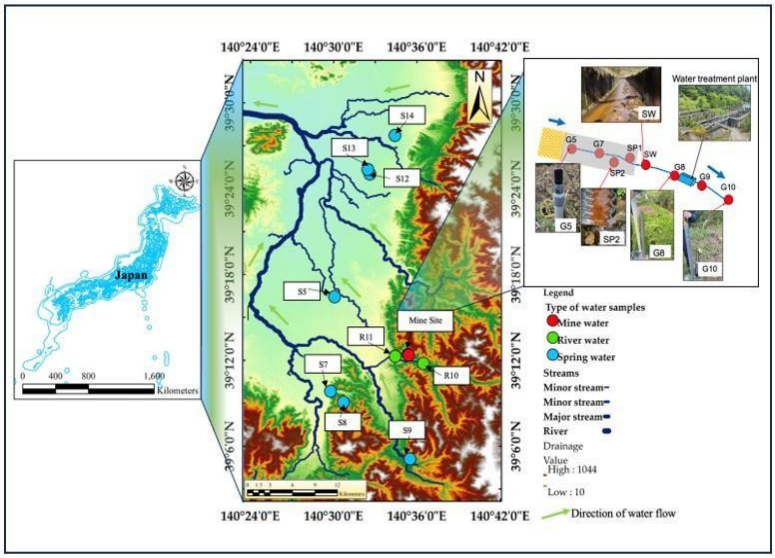

Fig.1 Location of the study area including the mine site and its nearby river and spring water resources.

2. Impact of effluents from a closed mine on the surrounding water resources: Groundwater contamination of the historical abandoned mine site in Japan

Firstly, chapter 2 focuses on the geochemical characterization of mine effluents at the abandoned mine site and its effects on nearby water resources. In the mine site, the mine drainage from the adit, shaft, tailing dam, and sedimentation impoundments were transported to mine water treatment facilities downstream of the mine site whereas the details of the effluent channels and old mine adit are unknown. However, in the high snowmelt season, the increase in mine effluents occurs, and the effects of seasonal variations on groundwater geochemistry still need to be evaluated. Therefore, the geochemical characterization of groundwater, seepage, and surface water at the mine site and its influences on nearby water resources are necessary to establish AMD remediation strategies at the mine site. The affected area by AMD is delineated by characterizing the spatial and temporal distribution of PTMs, major cations, anions, and physicochemical parameters of mine water samples, locally famous spring water, and river water samples. The dominant hydrogechemical facies and their primary sources of influence were differentiated by using efficient statistical methods, including principal component analysis (PCA) and hierarchical cluster analysis.

From the mine site, groundwater, seepage and surface water samples from the mine site and spring and river water samples were collected in four different seasons: spring, summer, autumn, and winter. In total, 18 representative water samples were obtained (Fig.1). Groundwater (G5, G7, G8, G9, G10) and seepage water (SP1, SP2) samples were collected along the upstream to downstream, surface water sample (SW) before entering the mine water treatment plant located in downstream of the impoundment. Since the river is located downstream of

the mine site, the treated mine water is adjoined into the river. Therefore, the river water samples (R10, R11) were collected from the upstream and downstream (Fig.1). Locally famous spring water resources (S5, S7, S8, S9, S12, S13, and S14) were also collected along the drainage basin. The collected samples were brought to the laboratory for the analysis of major cations, such as Ca, Mg, Na, and K, and trace elements, such as Al, Cu, Fe, Pb, and Zn using an inductively coupled plasma atomic emission spectroscopy (ICP-AES; ICPE-9000, Shimadzu corporation, Kyoto, Japan), anion by ion chromatography (Dionex Integration RFIC System, Thermo Fisher Scientific K. K., Tokyo, Japan). Principal components analysis (PCA) was applied utilizing major cations, major anions and heavy metals concentrations of the water samples to identify correlations of the measured physicochemical/hydrogeochemical parameters of the samples (Glascock et al., 2007; Hasan et al., 2020; Mufalo et al., 2021; Rahman et al., 2022; Soro et al., 2023; Tomiyama et al., 2019). Data analysis was performed using OriginPro (OriginLab Corporation, Northampton, MA, USA). Thermodynamic speciation modelling for the water samples was conducted using PHREEQC code with the minteq4 database (Parkhurst & Appelo, 2013) and Eh-pH stability diagrams were created using the Geochemist's Workbench® (Bethke, 1992).

According to the result of seasonal investigation, the mine water samples showed the typical AMD characteristics; Ca^{2+}-SO_4^{2-}/ Mg^{2+}-SO_4^{2-} type resulted from the oxidation of sulfide-bearing minerals from the tailing dam, sedimentation impoundment, mine adit, and old mine working facilities (Fig.2a). The mine water samples are significantly contaminated by Zn, Al, Fe, Cu and Pb, which are higher than the Japan effluent standards. However, in the vicinity of the mine site, the water resources of the river water (Ca^{2+}-HCO_3^-, or Ca^{2+}-SO_4^{2-} type) and spring water samples (Ca^{2+}-HCO_3^- or Na^+-K^+-HCO_3^- type) had neutral to alkaline pH, showing no AMD influence (Fig.2b). This implies the effectiveness of the water treatment plant at the mine site, the higher dilution capacity by river water, and the abundance of neutralization minerals in spring and river water resources. This infers that the 300-years old mine still has the potential to produce AMD. In addition, the groundwater samples downstream of the mine site also showed the influence of AMD and contaminant transport. Therefore, the changes in geochemical properties through historical and current AMD highlight the evolution of AMD over time and the possibility of the natural attenuation mechanisms at the mine site. The abundance of toxic elements at the mine site is affected by the mineralogy of Kuroko's ore deposit and the secondary minerals, mainly ferrihydrite, anglesite, and jarosite.

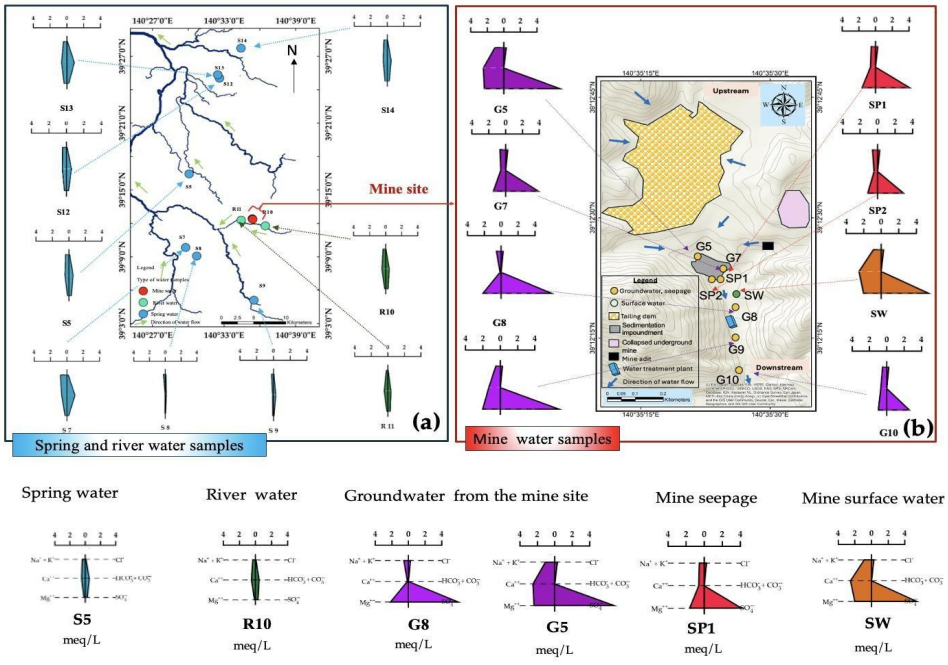

Fig.2 Stiff diagram for comparison of water qualities (a) spring and river water samples (b) mine water resources; groundwater, seepage, and surface water samples.

The PCA analysis and the cluster analysis could also effectively differentiated the AMD at the mine site from the uncontaminated water resources. The upstream groundwater, G5, downstream groundwater, G8 and G9, and surface water, SP1 and SP2, were positively correlated with PCA1, inferring the same sources of contamination

(Fig.3). The PCA1 showed loading of 37.6% for the spring water samples (S7, S12, S13, and S14) and river water sample (R11) downstream and seemed to be hydrogeologically related to each other (Fig.4). The results suggest the spring water and river water around the mine site are not influenced by the mine water. This indicates that the mine site does not have a significant influence on the spring and river water samples. Even after 300 years of the mine's existence, the site continues to release AMD with low pH. However, no contaminant transport occurs in the spring and river water. Additionally, the treatment plant installed at the mine site is effective in treating AMD.

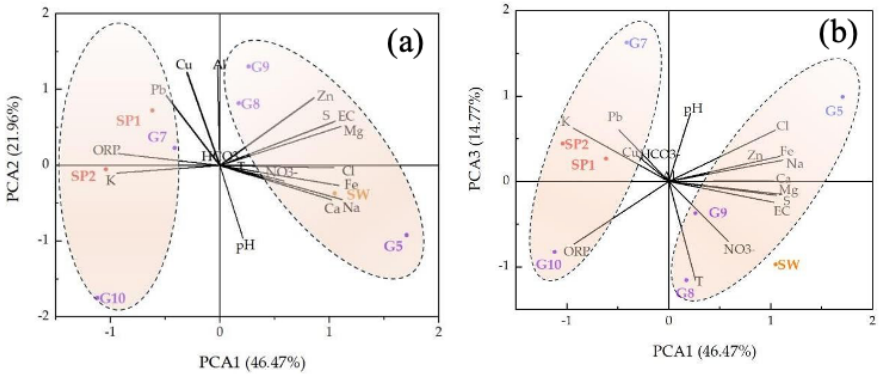

Fig.3 Principal component analysis (PCA) of mine water samples (November 2022). (a) PCA 1 vs PCA 2 with their respective loadings, (b) PCA 1 vs PCA 3 with prominent parameters.

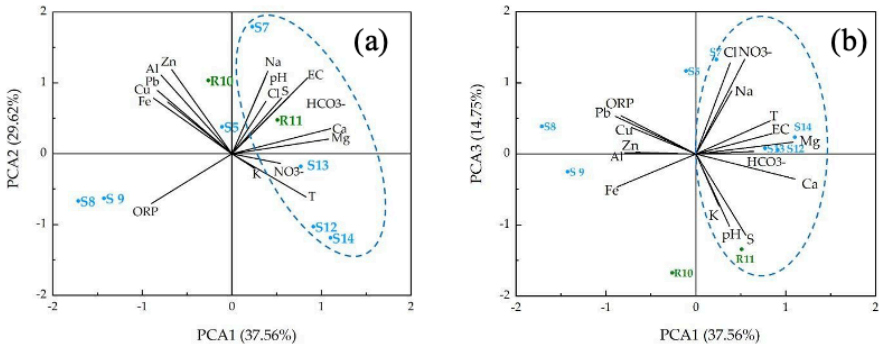

Fig.4 Principal component analysis (PCA) of spring and river water samples (November 2022). (a) PCA1 vs PCA2 with their respective loadings, (b) PCA1 vs PCA3 with prominent parameters.

The findings in this study imply that the identified contaminated water resources should be monitored and reclaimed. Thus, by considering the spatial distribution of contaminants reasonable countermeasures, and alternative strategies should be applied. For effective groundwater remediation, pumping the contaminated groundwater and treating it with a conventional water treatment plant is preferred. Another alternative option is installing a reactive permeable barrier or applying eco-friendly neutralizers in the downstream part of the mine site.

3. Environmental forensics of a 300-year-old legacy mine in Japan: Insight into the evolution of mine water quality and fate of contaminant transport

After the delineation of the effect of AMD from the mine site to nearby environment, chapter 3 evaluated the evolution of mine water quality from the 300-year-old legacy mine site and its sources of AMD from the tailing dam, mine adit, sedimentation impoundment and collapsed underground mine. Four drilled bore core samples and additional mine water samples were collected from upstream to downstream of the mine site. The mineralogical and chemical properties, as well as the qualitative and quantitative analysis of PTMS, were analyzed. These evaluations provide a valuable insight into the understanding of the leaching behaviours of PTMs, their speciation, and mobilization affinity within different geological media. The soil samples were provided for batch leaching tests by Japanese standardized method (Tabelin et al., 2014), and sequential extraction experiment by (Treaser et al., 1979). The resulted data were analyzed by PCA and well-known hydrogeochemical platform PHREEQC to understand the dominant geochemical processes.

From the result, regardless of the location of the borehole cores, all of these core samples are mainly

composed of quartz SiO_2 up to 82.6% and aluminosilicate minerals Al_2O_3 of up to 16.84 wt%, which are known to be major rock-forming minerals. Regarding trace metals, tailing borehole A contains the significant amounts of Zn ranging from 210 to 12,900 mg/kg, followed by Cu (78.1-7,400 mg/kg) and Pb (30.2- 1,800 mg/kg) exceeding the allowable limit of Japanese background soil (Shikazono et al., 2008). This observation can be linked with the long-term leaching behaviours of all core samples.

When focusing particularly on the tailing dam, apart from the topsoil and base rock, acidic pH is primarily observed in the middle part of the tailing, where the highest oxygen ingress (582-658 mV) favours the extensive oxidation of primary sulfide minerals along the depths of 0.10-16.0 m. This is evident by the relic of rhomboidal pyrite near the upper part of the tailing profile (2.4-2.6 m), resulting in relatively higher concentrations of dissolved metals, especially Cu, which reaches up to 4,400 µg/L, and Pb, which reaches up to 1,300 µg/L (Fig.5a). In contrast, Zn is likely to dissolve first and leach in the earlier stages of AMD evolution, resulting in lower leachable concentrations (900 µg/L). Approaching the middle part of the tailing dam, the lowest acidic pH, still an oxidizing condition, the transition of unsaturated-saturated zone appears (GL-10m) was observed. Under these acidic conditions, the disintegration of aluminosilicate and geogenic aquifer minerals, as well as the tailings gangue minerals, can occur, resulting in the dissolution of these minerals and the production of highly concentrated Al (14,700 µg/L) and Fe (11,000 µg/L) concentrations. This implies that Zn is the most mobile species, showing persistent liberation regardless of depth, while Cu and Pb are more stable, by forming secondary carbonates, or Fe-Mn oxidizes, which might inhibit their mobilization to the downstream groundwater.

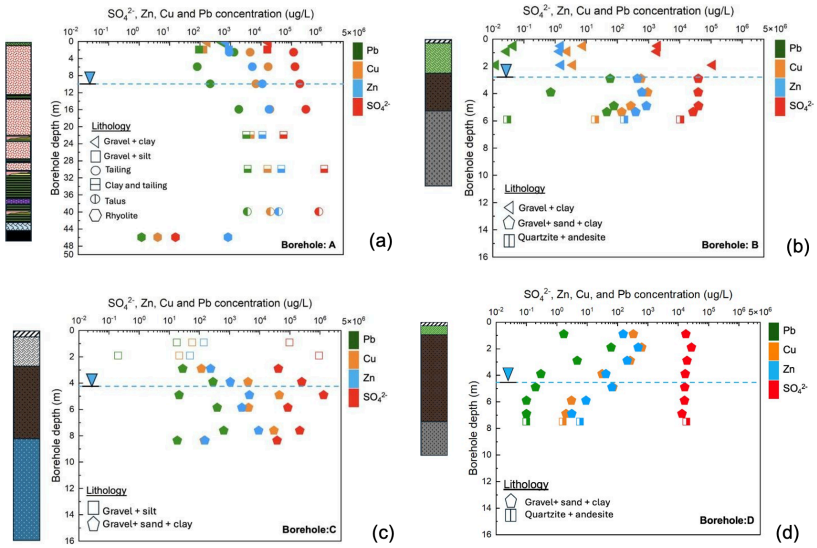

Fig. 5 Leaching behaviours of soil samples and their respective leaching behaviours for Zn, Cu, and Pb, with their SO_4^{2-} concentrations. (a) Tailing borehole, A, (b) borehole B, (c) borehole C, and (d) borehole D.

In addition to the result of long-term leaching behaviors, solid-phase partitioning of core samples in the tailing dam also inferred that acid leachates will still be released for decades or years or more. In the tailing dam, Zn is found to be the most mobile species, with an almost equal distribution across four phases: carbonate (0.28-35.1%), Fe-Mn oxides (16.8-30.6%), sulfides/organic matter (1.79-30.9%), and exchangeable (3.71-19.3%) phases. Zn has the highest exchangeable fraction compared with Cu and Pb in the tailing (Fig. 6a). This implies that Zn is weakly adsorbed to the soil and is readily released into the environment, posing an ecotoxicological risk (Concas et al., 2006; Romero et al., 2006; Shikazono et al., 2008; Van Geen et al., 1997). After Zn, Cu is found to be the second most mobile species in the tailings, which are mainly associated with primary sulfide minerals that remain after the mineral processing mainly distributed in the sulfide phase (6.85-36.6%) and the exchangeable phase (1.62-31.6%), while the carbonate phase was negligible (Fig. 6b). In nature, Pb is known for its higher adsorption affinity to soil and relatively slow mobility compared with groundwater flow (Fig. 6c). In AMD affected areas, the formation of secondary lead minerals might restrict the mobility of Pb. Still, although Cu and Zn pose an immediate ecological risk, the remobilization of Pb should not be underestimated due to its significant environmental threats to human health (Dang et al., 2002; Galán et al., 2003; Liang and Thomson,

2009; Romero et al., 2006). Another important factor is that the gradual decrease in total metal contents in the downstream of the mine site implies the anthropogenic impacts of the mine site. However, both the batch leaching and sequential extraction showed the possibility of toxic metal adsorption under neutral or alkaline pH and the dilution with the lateral the inflow of upstream natural water resources.

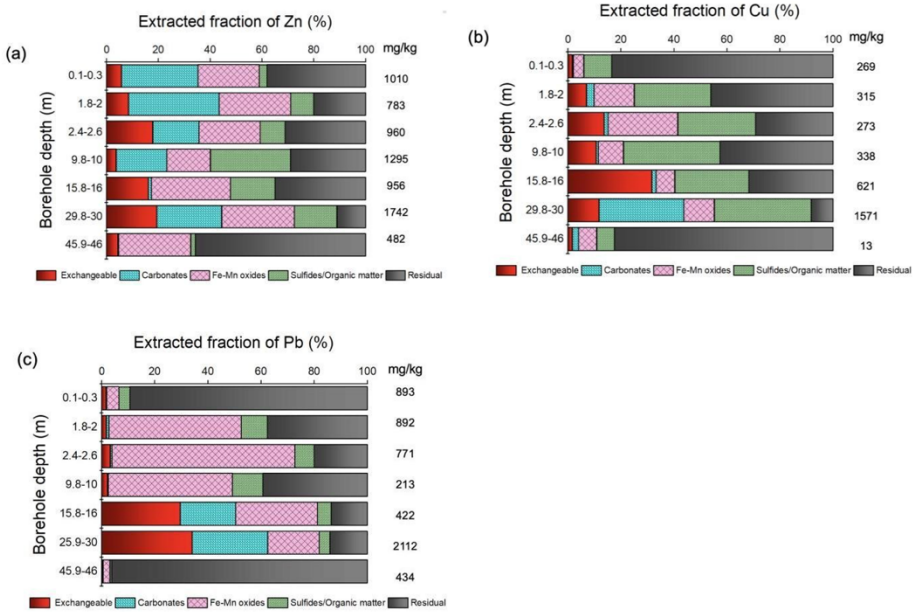

Fig. 6 Solid-phase partitioning of tailing borehole A, (a) Extracted fractions of Zn, (b) extracted fractions of Cu, and (c) extracted fractions of Pb.

This chapter evaluated the sources of AMD and its geochemical control on the evolution of mine water quality. Meanwhile, the oxidation of sulfide minerals in the downstream of the mine site, and deeper groundwater infiltration from the mine adit, sum up the additional anthropogenic input in the downstream. Therefore, although the shallow water resources are treated by a water treatment plant, the challenge arises from the infiltration of contaminated deeper groundwater resources from the mine adit to the middle of the mine site. However, the geochemical characterization of water and soil samples, and statistical analysis also provide the evidence that dilution, buffering, and natural attenuation may occur downstream of the mine site (borehole D). This can be expected due to its higher fractionation with Fe-Mn oxides and their concentrations (XRF, XRD), as well as a decrease in their total extractable phases with increasing distance to the downstream. Therefore, this study provides important insights that immediate remediation is not yet needed.

4. Transformative prediction of the groundwater migration and its contaminant transport: a multidisciplinary approach coupling transport mechanisms and radioactive dating

In chapter 4, the mine site's historical hydro-chronology, time series plume migration, and its future environmental risks were estimated using the conservative environmental tracer (^{3}H). Various sulfide minerals, including those in the tailing dam, sedimentation impoundment, mine adit, and the collapsed underground mine, are responsible for the formation of AMD. Subsequently, these leachates can percolate through advectional dispersion and contaminate the soil (aquifer) and groundwater resources. This implies the formation of the leachate plume, which has the potential to migrate downstream of the mine site over time. Due to these perspectives, this chapter proposes a constructive methodology for sustainable environmental management, which is feasibly applicable worldwide. Due to the hydrologic complexity of the mine site, the contaminant migration along with groundwater flow is the main objective of this chapter. The approach involves reconstructing historic groundwater using an environmental tracer (^{3}H), developing the migration pattern of groundwater velocity and its retention time, and predicting time-series-dependent plume migration.

When contaminant groundwater passes through the soil/sediments, the metals Zn, Cu, and Pb behave differently, giving rise to varying transport rates relative to the groundwater flow velocity (Seidou et al., 2022). The soil-water-

contaminant system encompasses geochemical dissolution, precipitation, and sorption processes, as well as physical aspects include advection, dispersion, and molecular diffusion phenomena. These phenomena are commonly described as distribution coefficient (K_d), and retardation factor (R_f) (Allison & Allison, 2005). This method can be applied to lab-scale experiments, including batch method, flow-through column method, field modeling and situ batch methods. This approach is widely used in hydrogeological studies, especially contaminant transport in anthropogenic disposal sites, such as mine sites' tailing storage facilities (TSF), industrial and radioactive wastes (Twining et al., 2004).

In the previous investigations, we observed the downstream groundwater contamination from the 300-year-old legacy mine site. These valuable findings were achieved by developing a constructive methodology that integrated by reconstructing the hydrological signatures of groundwater samples, lab-scale solid-phase partitioning and solid-liquid partitioning methods to determine (K_d) and (R_f). Based on the results, the lifetime of the contaminants' migration and their reach to the river can be estimated. In the natural environment, the sulfide minerals (pyrite, galena, and sphalerite) exhibit varying solubilities and geochemical affinities (Elghali et al., 2023). The metals (Zn, Cu, and Pb) tend to fractionate with freely mobile phase, carbonate, Fe-Mn oxides, sulfides/organic matter, and the crystal lattice of the minerals (Heuel-Fabianek, 2014). To reconstruct the contaminant migration patterns, six representative groundwater samples were collected in 1-litre polyethylene containers at the following locations: the mine site's tailing dam (G1), the mine adit (G4), the sedimentation impoundment (G5 and G7), and downstream groundwater (G8 and G9). These collected water samples were sent to Geoscience Laboratory in Aichi, Japan. The analysis followed the "Radiation Measurement method series. No.9: Tritium Analysis Method" (Revised October 2023, Nuclear Regulation Authority, Japan), using the electrolytic enrichment–liquid scintillation counting method.

From the result, hydrochronological investigation delineates the sources of groundwater from the tailing dam and mine adit, indicating a groundwater age of less than 50 years, derived from recent recharge (Fig.7). The middle and lower parts of the mine site show the mixing of older and younger resources, which is attributed to the linear downstream groundwater flow. The accelerated groundwater velocity in the downstream gradient of sedimentation impoundment stresses the influx of contaminants, which are then transported downstream. Zn exhibits the highest mobilization affinity in conjunction with groundwater flow, in contrast to Cu and Pb. Therefore, from the mine site, Zn is the most mobile metal and is loosely bound to the soil, while Pb exhibits the highest binding strength to the soil through complexation with Fe-Mn oxides. Based on the rates of contaminant transport and groundwater flow, the frontal plume of Zn reaches approximately 500 m, while Cu and Pb only reach 313 m. Therefore, it can be inferred that the secondary minerals can act as effective scavengers in the aquifer downstream, serving as a natural barrier that hinders the release of Pb into the groundwater.

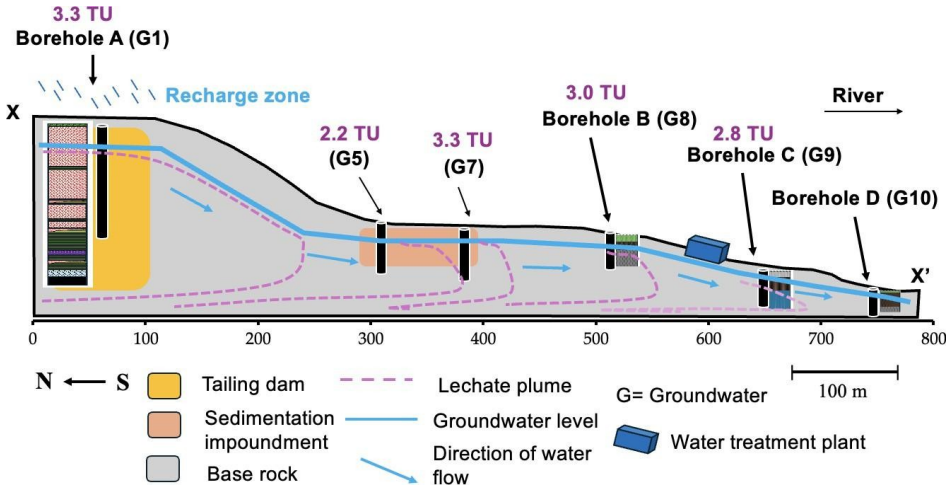

Fig. 7 Cross-sectional profile of the mine site showing the groundwater hydrochronology by using the environmental tracer ^{3}H.

By applying the historical contaminant transport, the migration time of each contaminant (Zn, Cu, and Pb) along the downstream direction can be predicted, facilitating the estimation of future groundwater

environmental risks. As illustrated in Figs. 8a and 8b Zn shows the shortest travel time from the sedimentation impoundment, taking within 415-1,050 years to travel downstream along the rapid groundwater flow (121 m/year), while Cu taking 2,120-5,370 years and Pb requiring over a longer travel time of 10,900-27,700 years, respectively. Based on the developed time series plume migration, the potential risks of its travel time to the river can be predicted. It is expected that Zn will take approximately 1,860 years to the river, while Cu and Pb will take 9,460 years and 48,900 years, respectively. Thus, we still have enough time to select appropriate, reasonable countermeasures.

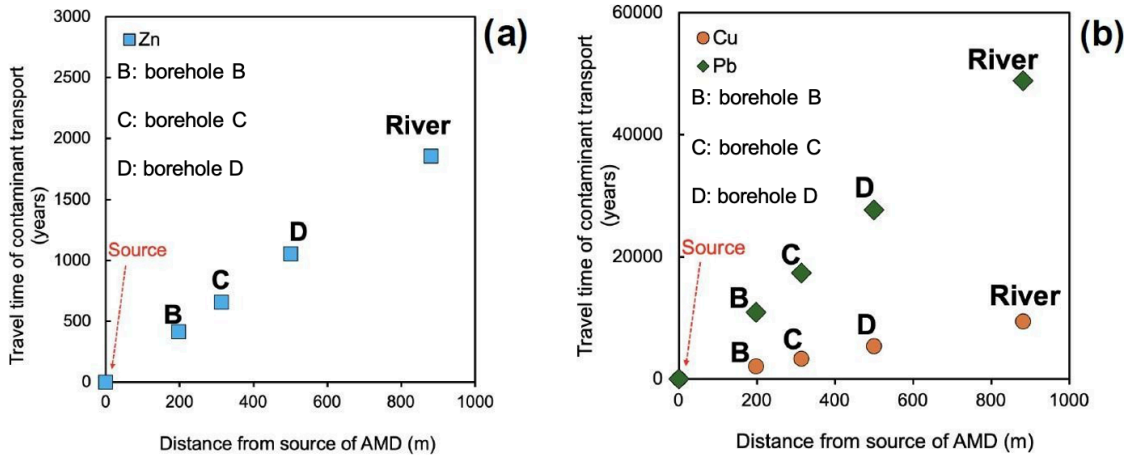

Fig. 8 Estimation of the contaminant migration time or plume travel time (years) of the mine site to the river. (a) Migration of Zn to the downstream of mine site, (b) migration of Cu and Pb to the downstream of the mine site. (B, C, D refers to the location of the borehole C, borehole C, and borehole D).

5. Conclusion

There are hundreds to thousands of legacy mine sites, and among them, a significant number of mine sites already induce AMD. Due to the unavoidable and prominent effects of AMD, numerous proposed approaches for sustainable management have been proposed in every part of the world. However, there are still challenges regarding groundwater contamination in areas with historical legacy mines, which have complex and unforeseen contaminant pathways, and limited historical records. This is making it challenging to develop effective long-term strategies for addressing AMD. Most of the challenges in legacy mine management are complex, unforeseen contaminant pathways. There are various applications, such as geophysics and seismic transmissivity approaches; however, these approaches are economically demanding evaluation. Additionally, these approaches are not suitable for the current complex site conditions. The prediction of AMD formation using a geochemical approach, such as acid-base accounting, is not appropriate because AMD already exits, and its trend of evolution could be understood by monitoring historical surface water contamination. however, the problem persists with groundwater contamination.

This study assessed the long-term environmental impacts of anthropogenic mining activities on surface and groundwater resources, as well as the future mine site management by selecting one of the 300-year-old legacy mine sites in northern Japan. The research objectives were achieved through a constructive methodology approach, involving both field investigations and laboratory experiments. In this dissertation, each objective and relevant finding is systematically outlined in five chapters including general conclusion. Overall, the methodology in this research includes: hydrogeochemical characterization of collected water samples, chemical and mineralogical analysis (XRF-XRD), SEM-EDS, standard batch leaching tests, and solid-phase partitioning. Additionally, the environmental tracer (^{3}H) is used to determine the mine site's hydrological age, predict contaminant transport rates, and finally purpose suitable long-term AMD remediation strategies.

To overcome these problems, this research proposes a constructed methodology primarily comprising six parts. The first part involves assessing the large-scale area, where complex hydrological systems complicate the understanding of the effects of AMD. Firstly, the legacy mine sites and their impact on nearby environments, as well as delineating AMD-contaminated areas through effective hydro-geochemical characterization. In differentiating between AMD-contaminated and non-contaminated areas, the statistical approaches of PCA and dendrogram could be effectively applied. After that, the second approach focuses on the mobility of contaminants and their retention mechanisms in geological media and groundwater in the contaminated area. These can be evaluated by mineralogical

and geological investigations (leaching and solid-phase partitioning). This approach provides the critical framework of standpoints, the potential of acidification or neutralization in the geological media, the extent of contaminant migration plumes along the groundwater, and the long-term contaminant release from sources of AMD to nearby environment. These will provide insightful outcomes for further AMD remediation strategies in anthropogenically contaminated sites. The main challenge in the estimation of the fate of contaminant transport can be overcome by integrating a universal approach of environmental tracer. This approach is further extended by theoretically adapting the solid-liquid partitioning to develop contaminated hydrology, contaminant migration and groundwater flow. These three combined steps of approaches, including the environmental tracer ^{3}H. These could successfully achieve the following steps (5), and (6) that is to reconstruct the historic contaminant pathways, prediction of the fate of groundwater flow, which is the prominent agent in contaminant transport and estimation of the contaminant migration to the nearby water resources.

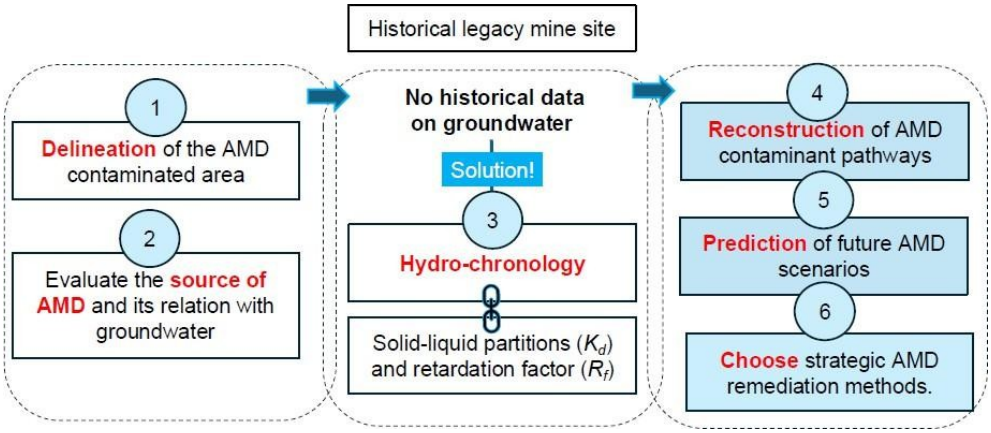

Fig.9 The flow chart of the proposed constructed methodology for long-term AMD management.

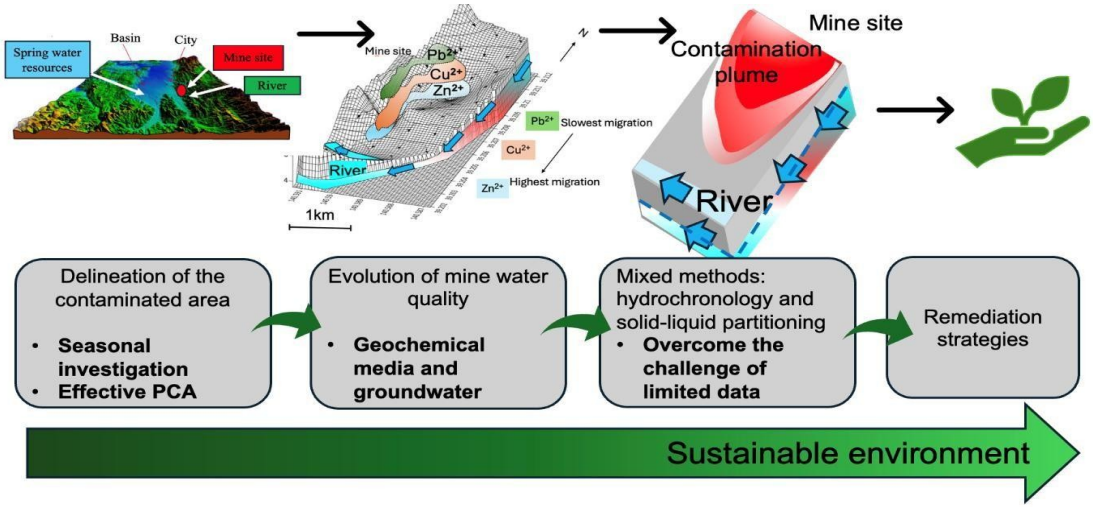

Fig.10 The conceptual diagram for the implications of the study in obtaining the sustainable management.