研究主論文抄録

論文題目 Spatio temporal Modeling and Identifying Mechanism of Groundwater
Arsenic Contamination in a Hydrothermal Mineralization Area

(熱水鉱化域における地下水ヒ素汚染の時空間モデリングと汚染メカニズムの特定)

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主論文要旨

《本文》

Arsenic (As) is one of the most hazardous elements for human health, causing carcinogenic skin diseases, internal cancers and non-carcinogenic diseases such as diabetes, peripheral neuropathy, and cardiovascular conditions. A study site in North Sulawesi, Sulawesi Island, Indonesia was selected as it was suitable for investigating the importance of fault lines, geothermal and hydrothermal mineralization to groundwater As contamination. Stable isotope analysis was used to ascertain the groundwater source and the mixing mechanism of the shallow and deep groundwater. Geostatistical modeling by ordinary kriging revealed consistent general patterns of As concentrations during the past ten years, with high concentrations found along a NW-SE axis. Co-kriging method were used to map As spatial pattern in mineralized sedimentary rocks area which in this rocks type As in rocks and As in groundwater has good positive correlation. Those methods are validated with microscopic, spectroscopic and scanning electron microscope (SEM) analyses of rock samples for mineral identification to understand the interaction between water and rocks.

Ordinary kriging estimates for ten years (1995-2004) clarified that the spatial patterns of As concentrations differed significantly from the other three species. The most remarkable features were that the location and direction of the regions with high As concentrations were almost the same over the ten years, and that these regions corresponded with the spatial pattern of low δ^{18} O along NW-SE fault zone. By

comparing the advection dispersion equation simulation results for the two presumed As sources (the mine and the fault zone) with the ordinary kriging estimates of the As concentrations, it was determined that the deep-seated geothermal system and the rise of hydrothermal fluids through the fault zone are probably the main As sources in the shallow groundwater. The area contaminated by the As-rich mine waste water was found to be small.

Jarosite was identified as a main fill mineral in cracks and pores of the mineralized sedimentary rocks. The most plausible origin of the jarosite was the oxidation and leaching of As-rich pyrite. This oxidation/leaching and abundance of Fe-sulfide rich minerals in the mineralized sedimentary rocks may be the reason for the high correlation between As concentrations in groundwater and in the rocks.

The co-kriging result identified local anomalies in groundwater As concentrations over the permissible limit (10 ppb). Such anomalies did not appear through ordinary kriging. Co-kriging allowed characterization of the great variability in groundwater As concentrations. There are two possible causes of groundwater As contamination, including characteristics of the fractured aquifer as well as the difference in mixing of geothermal fluids with high As concentrations from depth with shallow groundwater. The first mechanism is attributable to groundwater mixing from different reaction zones, different fracture systems, and preferred flow directions in a fractured aquifer, while the second is controlled by depth of the fractures which would determine the amount of As supplied in shallow groundwater.

SEM and EDS study include element mapping techniques strengthen microscopy and spectroscopy observations. Jarosite was found as cubic minerals among calcite and silica mineral as major mineral for fractures fill mineral. The presence of the mineral also signed by Fe-S-O elements from EDS element map results. Based on pH-Eh condition of discharge water from, jarosite from both samples appear in unstable conditions and possible changes to other forms such as goethite. Accumulation of As elements along with the Fe-S elements in mineralized rock sample is indicating jarosite is alteration of As-rich pyrite or arsenopyrite. Different result from host rock of hot spring discharge, As elements are found to be scattered and do not co-located with Fe-As elements. These findings also indicate existence of two different mechanisms which it is interpreted that jarosite in mineralized rock is related with oxidation of As rich pyrite.