Japan Geoscience Union Meeting 2012
(May 20-25 2012 at Makuhari, Chiba, Japan)

AHW30-P16 Room:Convention Hall Time:May 22 17:15-18:45

Formation process of Arsenic contaminated groundwater at recharge area in Sonargaon, Bangladesh

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Ganges delta plain is the largest affected area of As contaminated groundwater in the world. Since the As contaminated groundwater occurs mostly in the reducing aquifer condition in this area, the formation mechanism has been believed that the As adsorbed onto Fe-oxyhydroxides/oxides was released in association with reduction-dissolution of the host phase via microbial activity. However, in Sonargaon, located east from Dhaka in Bangladesh, the As contaminated groundwater occurs in the actively recharging and oxic zone, and the conventional mechanism cannot explain the case. In this area, a host phase of As is chlorite. Based on the chemistry of groundwaters taken from -5, -10 and -15 m test tube wells (total 12 wells) and about 50 household tube wells drilled in the actively recharging zone, we documented that the As was released between -5 and -10 m depth via chemical weathering of chlorite. In this area, the groundwater at -5 m contained ˜0.4 mg/L As and 0.8 mg/L at -10 to -15 m, and >1.1 mg/L at -24 m (Maeda et al., 2011). In order to quantitatively argue the contribution of chemical weathering of detrital mineral(s) to the As and related chemical composition of As contaminated groundwater, REE elements of groundwaters and detrital plagioclase and chlorite picked from the contaminated aquifer sediments were analyzed using ICP-MS. Plagioclase is one of the most soluble mineral during chemical weathering and cholorite was the most plausible candidate of As source of the contaminated groundwater of this study. The obtained REE data was normalized using the data set of shale.

All REE paterns of the studied groundwater give the positive Eu anomaly, and most of those the negative Ce anomaly, which becomes larger with increasing depth. Groundwaters taken from test tube wells give the increasing As concentration with increasing concentrations of heavy REEs. REE concentrations of the chlorite are higher than those of the plagioclase, and the normalized concentrations are almost the same among the REEs. While, the plagioclase enriches in Eu, and the light REEs are enriched compared with the heavy REEs. As contaminated groundwaters of this area has Ca-(Mg)-HCO3 type major composition. Calcium and Mg are the major elements of plagioclase and chlorite. REE analysis confirmed that the chemical weathering of those minerals control even the major water chemistry.

Positive Eu and negative Ce anomalies do not have good correlation to the As concentration of the groundwaters. The positive Eu anomaly indicates active dissolution of plagioclase in the aquifer, while the dissolution of this mineral does not cause As contamination. Ce is easily oxidized in oxic aqueous condition from III to IV valance to be removed from the solution due to precipitation as oxide. Thus, the negative Ce anomaly indicates the reduction of groundwater with increasing depth of the aquifer. No relationship between As concentration and degree of negative Ce anomaly suggest that the As release into this groundwater aquifer does not occur in association with the reduction. In conclusion, the REE patterns of groundwaters and detrital minerals support our hypothesis that the As is released from congruent dissolution of detrital chlorite via chemical weathering.

Beneath the studied area, Holocene As contaminated aquifer and Pleistocene uncontaminated aquifer separated by the intercalated impermeable clay layer are present. However, the clay layer lacks just beneath the highest As contaminated well studied here, and the two aquifers directly contact to each other. Excess use of deeper groundwater for irrigation would promote accelerated vertical infiltration of oxic surface water into the shallow aquifer, and the appearance of oxic condition in the groundwater must cause As release from the chlorite via chemical weathering.

Keywords: arsenic contamination, groundwater, Bangladesh, Chlorite, weathering, rare earth element