Historical Cu slags of the Rudawy Janowickie Mountains (SW Poland) – Mineralogy and potential environmental impact

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Historical mining and smelting areas give opportunity to study long-term mobility of potentially toxic elements (eg. As, Cu, Zn and Pb) and their interactions with soils and surface waters.

The Rudawy Janowickie Mountains were an important centre of copper mining and smelting in Poland until 1925 with metallurgical activities being documented as early as in XIV cenury. The exploitation of Cu ores left large amounts of mine tailings and slags covering the surface area of around 35ha.

Studied Cu slags were sampled in the vicinity of two streams and from streambeds. Slags are isometric and well rounded, with size ranging from few to dozens of centimetres. Dominant type of slag is black, massive and displays flow textures. Other slag type is highly porous and often encloses small fragments of granitic rocks. The average chemical composition of slags is 46.8 wt. % Fe_2O_3, 40.0 wt. % SiO_2, 8.1 wt. % Al_2O_3. Slags show elevated concentrations of Cu (above 1 wt. %), Zn (up to 3644 ppm), Pb (up to 268 ppm) and As (up to 131 ppm). The slags consist of silicate glass, fayalite, hercynite, bornite, pyrhotite, intermetallic compounds of Fe and As, metallic Pb and Cu. The slags have been subjected to weathering and observed secondary phases are represented by brochantite and Fe oxy-hydroxides. The weathering products occur as aureoles and interstitial replacements within the sulfides – the most important metal and metalloids carriers.

Our observations show that studied slags undergo weathering processes and release potentially toxic elements (especially Cu). The presence of secondary phases, especially highly soluble Cu-sulfates (brochantite) may imply that the elements are highly mobile and contaminate water, soils and sediments. It is confirmed by preeliminary geochemical analysis of stream sediments. The Cu concentration in sediments from the area where no slags have been found is 8 ppm, while in the site where slags are widespread Cu concentration exceeds 130 ppm.

Depleted uranium found in Japanese reference fallout material for activity measurements

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As well known, uranium isotope ratios do not change substantially in the nature. The ratio that is different from the natural ratio is thus resulted from anthropogenic nuclear activities except from natural nuclear reactors. We have determined the $^{235}U/^{238}U$ ratios in four lots of the reference fallout material for activity measurements (RFM) prepared by Japanese Meteorological Research Institute (MRI). The RFM is a mixture of monthly atmospheric deposits collected at 14 monitoring stations sited all over the Japanese Islands between 1963 and 1979. All the lots of the RFM that we analyzed show obviously low $^{235}U/^{238}U$ ratios, around 0.0071, in comparison with the natural ratio, 0.00725. In addition, the ratio in the acid soluble fraction of the RFM is lower than that in the acid insoluble fraction. These analytical results suggest that the RFM is contaminated with uranium having a low $^{235}U/^{238}U$ ratio, i.e., depleted uranium (DU). If the $^{235}U/^{238}U$ ratio of DU comprised in the RFM is 0.002, percentage of DU to the gross uranium amount in the RFM is roughly estimated at 2%.

Warneke *et al*. [1] suggested the possibility of fallout of DU in UK in 1977, 1978 and 1983 based on their analytical results of the uranium isotope ratios in the archived herbage samples of the IACR-Rothamsted. If DU of the RFM has the same origin as that of DU found in the herbage samples, DU might have fallen as a global fallout in the late 1970s. In Japan, the deposition of anthropogenic radionuclides originated from nuclear test explosions predominate in the west side of the northern Japanese Islands, especially in Akita. The $^{235}U/^{238}U$ ratios in the atmospheric depositions collected at Akita in March in 1977 and 1978, however, are nearly the same as or only slightly higher than the natural ratio.

It is in no doubt that the RFM is contaminated with DU. DU must have fallen somewhere in the Japanese Islands in the 1960s and/or 1970s. The next step of the research is to identify the origin of DU in the RFM.

[1] Warneke *et al.* (2002), *EPSL* **203**, 1047-1057.