Mobilisation of Arsenic in Mine Tailings Subjected to Spontaneous Combustion

Jean-Marie Ferreux (ferreux@excite.com)¹, Marc Steinmann (marc.steinmann@univ-fcomte.fr)¹, Catherine Bertrand (catherine.bertrand@univ-fcomte.fr)¹, Claude Dubois (claude.dubois@univ-fcomte.fr)¹ & Yves Paquette²

¹ Université de Franche-Comté, Laboratoire de Microanalyses Nucléaires, 16 route de Gray, 25030 Besançon Cedex, France

² Institut national de l'environnement industriel et des risques - INERIS, c/o HBCM, 4 square François Margand, BP 534, 42007 St-Etienne Cedex 1, France

Introduction

Several open pit coal mines in France are actually being closed and transformed into agricultural and leisure areas. The transformed sites consist to a large extent of embankments of mine tailings, sometimes including lakes obtained by simple cessation of groundwater pumping in former open pits. The surface runoff of these sites will after complete restoration be introduced into the existing hydrological network. The mine tailings used for restoration are mainly consisting of bituminous shales, which are, like most organic-rich sediments, naturally enriched in arsenic and other heavy metals. These metals seem under certain conditions to become remobilised as suggested by As concentrations of up to 0.025mg/l found in surface and subsurface waters adjacent to sites in restoration. These values are above the recommended threshold level for drinking water of 0.01mg/l as defined by the US Public Health Service, but still remain below the maximum permissible limit of 0.05 mg/l proposed by the same organisation. The observed release of arsenic into the hydrological cycle may have serious consequences for water quality and public health. The scope of the present study is to ameliorate our understanding of the processes mobilising arsenic from mine tailings. To do so, two sites have been chosen in the northern (Montceau-les-mines) and southern part (Decazeville) of the French "Massif Central" where coal has been mined in Upper Carboniferous strata (Stephanian) since the beginning of the 19th century. Both sites are open pit mines located in direct vicinity to urbanised areas and in the final stage of exploitation. However, restoration has commenced in those parts of the sites where exploitation has been discontinued earlier.

Batch experiments

Our field data suggest that arsenic is released into the hydrological cycle essentially in areas where mine tailings have undergone spontaneous combustion. This process is a wellknown phenomenon in coal mining and triggered by slow oxidation of organic matter and pyrite under conditions not permitting ready dissipation of heat. In order to test this hypothesis and to get detailed information on the processes leading to As release, comparative batch experiments have been conducted on powder samples of unburnt and naturally burnt bituminous shales and mine tailings. Identical lithology and comparable mineralogical and chemical whole rock compositions were used as criteria to select burnt-unburnt sample pairs. 8g of sample powder were leached at 70°C with 80ml of demonised water (pH 7) in acid-cleaned PP containers at different extraction times. After leaching, the supernatants were separated from the powders by filtration (0.45 μ m) and analysed by liquid chromatography, AAS and ICP-MS.

Preliminary results and discussion

Preliminary results from a first extraction series are shown in Figure 1 below. Almost 3 times more As has been released by the burnt sample. In contrast, sulphate concentrations are much more elevated in the leachate of the unburnt sample. Our results thus clearly show that arsenic is more mobile after combustion, whereas sulphur is fixed more strongly, which is in agreement with the field data mentioned above. Correlations between arsenic and sulphur in unburnt whole rock samples suggest that As is prior to combustion fixed in a sulphurous mineral phase, most probably pyrite. Our present working hypothesis is that pyrite is oxidised during spontaneous combustion of mine tailings leading to the mobilisation of arsenic and sulphur. Arsenic remains mobile after decomposition of pyrite, whereas sulphur seems to precipitate as poorly soluble secondary mineral phase. Presently, additional batch experiments are conducted with acid and basic starting solutions in order to characterise As mobility over a larger pH range and to get further information on the solubility of the different mineral phases. Furthermore, we are trying to identify more precisely the different As carriers before and after combustion by SEM.



Figure 1: Results of batch experiments on burnt and unburnt mine tailings