Physical properties of sediments deposited in the minewater from a closed coal mine

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Abstract: The Chikuhou coalfield, situated close to the Onga River in the Chikuhou region, northern part of Fukuoka Prefecture, Kyushu Island, was once an important coal mining area in Japan. The colliery is now closed and waters derived from the mine flow into nearby river via springs. These 'spring waters' from the mine are highly coloured and locally called 'red water'. The mine water generally has high electrical conductivity and high concentrations of dissolved iron and sulphate ions. As a consequence, the water from this river is isolated and not used in the potable water supply or for irrigation but may be used in emergencies, such as fire fighting.

In this study, we have investigated the river sediments, focusing on the deposits from the red water. We focused on two localities, Sensui and Namazuta, which have different features. Water and suspended solids were collected at each outflow and where suspension occurred so called 'Akamizu'. Waters at Namazuta have higher pH and turbidity than at Sensui, which has higher acidity resulting in higher Fe ion content. We consider that the oxidizing bacillus in the mine waters is more active at Sensui as indicated by more red downstream and considerable 'Akamizu' pollutant. Therefore, it is thought over a long time period (semi-permanently) the oxidation of ferrous ions continues to exchange slowly and the value of electrical conductivity is generally about constant. At Namazuta, after rapid oxidation, the pollutants settle more slowly. Finally, 'Akamizu' has higher concentrations of cations and anions, derived from the mine waters, have flowed continuously into the Onga River since the mine closed.

Résumé: Le bassin houiller de Chikuhou situé près du Fleuve Onga, dans la région de Chikuhou du Préfecture de Fukuoka, Kyushu Island, tenait une fois une grande importance dans l'industrie houillère japonais. Les mines de houille sont abondonées actuellement et les eaux qui s'élèvent des mines se jetât dans la rivière par le moyeu des sources. Les eaux des sources sont plein de couleur et possèdent l'appel, dans le coin, de 'l'eau rouge'. Les eaux qui s'écroulent des mines on habituellement une conductivité électrique élèves, et ils contrôleuse aussi des concentrations très hautes de fer dissoudre et des ions de sulfate. Par conséquent, les eaux de cette rivière sont gardes en isolation des ressources générales de la région.

Keywords: acid mine drainage, water quality, coalmines, environmental geology, organic materials, properties.

INTRODUCTION

Industrial and domestic effluent has polluted the rivers of Japan. A great deal of effort has gone into controlling serious local industrial pollution improving river quality. Discharges from inadequate domestic drains and leaking sewers currently provide a majority of water pollution. Further improvement of river water quality will need reductions in other sources of pollutions, which are now being studied in detail (Ohishi et al. 2003).

Pollution from coalmine water is well known and is indicated by red water, so called 'Akamizu'. This is well known in one of the branches of the Onga River. This water is not used for domestics or agricultural purposes and is only used as an emergency supply such as for large fires. The pollution is seen mainly as a precipitate in suspension or deposited on the riverbed. The suspension has low pH and high electric conductivity. The pollution is produced by the oxidation of iron pyrites from the sedimentary rocks that contain thin layers of coal. Recent studies have focused on two 'Akamizu' sources related to mine water. Both are from upper streams of the Onga River; one from Sensui district, Nishikawa area, and the other from Namazuta district. According to Moribe, Okamoto & Sakamoto (2003) and Moribe et al. (2003), the two areas have different water qualities, which relates to the acceleration of the oxidation of iron by bacteria.

In this study we have investigated some properties of mine water flowing into the river sampled in Sensui, the town of Kurate town and Namazuta, in the city of Iizuka. We also compared these waters with those of the Onga River. The results showed that the water 'Akamizu' of Sensui and Namazuta have high concentration of Fe^{2+}, Fe^{3+}, and SO_4^{2-}. Fe^{2+} is oxidized and becomes Fe^{3+} and then precipitates as a reddish suspension. However, where pH is low, Fe^{3+} often remains in solution, as in these conditions the solubility of Fe^{3+} is higher. 'Akamizu' contains more cations and anions than the Onga River below the confluence of the polluted streams. Water quality has changes dramatically in the last 40 years with a large decrease in suspended solid (S.S.) after closure of the mine and large increases in bacteria form domestic sewage. The differences in water quality between the mine water and the precipitate are indicated by the pH. The effects of the mine water from the Chikuho Coalfield on the water quality of Onga River are estimated from laboratory experiments and local site survey results (Kobayashi et al., 2004a, 2004b).

LOCATION

The are of investigation are shown in Figure 1, which shows the location of the sampling points of mine water and on the tributaries of the Onga River, Fukuoka Prefecture, Northern part of Kyushu Island in Japan.

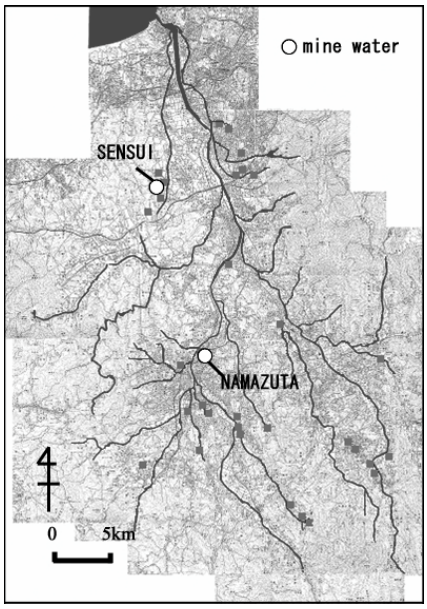

Figure 1. Sampling point of iron-containing precipitates and distribution of the mine water out flows in the Chikuhou coalfield (Kobayashi et.al., 2004a).

The areas geology is predominantly early Palaeogene sediments and includes coal from the Nohgata Formation. They comprise shale, mudstone, sandstone, conglomerate and thin coal layers.

METHOD

In this study we have regularly collected samples of water and suspension from the sample sites. Mineralogy of the suspension was determined by X-ray diffraction (XRD) and chemistry by X-ray fluorescence (XRF). Petrology of the suspension 'Akamizu' was described from Scanning Electron Microscopy (SEM) micrographs. The surface area was measure by the B.E.T. method. Pore size distribution curves of each pollutant as 'Akamizu' were obtained as the result from calculation with B.J.H. method automatically after using by Thermo Quest SORPTOMATIC 1990 SERIES.

Water chemistry, measured by ion chromatography included the cations NH_4^+, Na^+, K^+, Mg^{2+} and Ca^{2+}, and the anions, Cl^- and SO_4^{2-}. Temperature (Temp.), electrical conductivity (EC), turbidity, dissolved oxygen (DO) and total dissolved solids (TDS) were also measured. Other measurements were made as indicators of other sources of contamination and included suspended solids (SS), biological oxygen demand (BOD), and faecal coliforms.

ANNUAL WATER QUALITY CHANGES OF THE ONGA RIVER

Annual changes in BOD, EC, SS and faecal coliforms in the Onga River show that BOD indicates changes in the biological activity, EC has been constant, SS indicates the operation of the coal mine and ore washing, faecal coliforms indicates changes in industry and population changes in the area.

RESULTS

We have discussed some of the properties of the pollutants on the water quality of Onga River. The results of a more detailed study of two localities at Sensui and Namazuta are described below. Location maps of collecting water and suspension in this study at Sensui and Namazuta are shown as Figure 1. 'Akamizu' occurred at the mine water outflow and suspension sample points. The water quality results of the mine water are in Table 1. The data show that the water at Sensui is more polluted than that at Namazuta being more acidic and having higher conductivity, much higher total iron and sulphate concentrations and slightly lower dissolved oxygen. Iron ions are more soluble in low pH, hence the higher concentration at Sensui as compared to the low concentrations of iron ions at Namatuza where the water is nearly neutral.

Table 1. Water quality results of the two mine waters.

Water quality test parameter	Namazuta	Sensui
PH	6.0	3.5
EC (mS/m)	162	260
DO (mg/L)	4.1	3.1
Total Fe (mg/L)	11.6	119
Sulphate ion (mg/L)	526	1430
Flow rate (L/s)	20	10

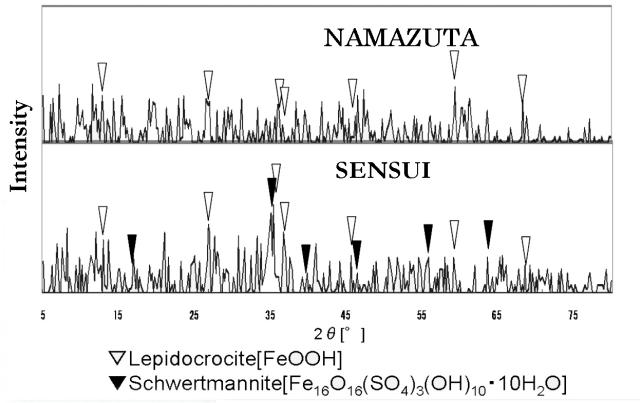

Figure 2. X-ray powder diffraction pattern of precipitates (upper) NAMAZUTA, (lower) SENSUI
▽Lepidocrocite [FeOOH]
▼Schwertmannite [$Fe_{16}O_{16}(SO_4)_3(OH)_{10}$•$10H_2O$]

X-ray diffraction patterns are shown in Figure 2. The material collected from Sensui typically contains minerals such as Schwertmannite ($Fe_8O_8(OH)_{8-2x}(SO_4)_x$) and Feroxyhite (FeO(OH), both of which are colloidal or have a low degrees of structural order (Nishida & Oku, 2002 and Sasaki & Tasaki 2001). Table 2 has the chemical composition of the precipitates. The precipitates from Sensui are composed mainly of iron and sulphur with minor silicon, aluminium and potassium, whereas the samples from Namazuta contain iron and silicon with minor amounts of aluminium.

Table 2. Chemical composition of precipitates in the mine water.

Sample	Al_2O_3	SiO_2	CaO	Fe_2O_3	P_2O_5	SO_3	MgO	MnO_2	Total
Namazuta	3.0	12.7	0.6	83.8	<0.1	<0.1	<0.1	<0.4	100
Sensui	2.7	1.8	<0.1	77.6	1.9	16	<0.1	<0.1	100

*Above data is described as % by weight.

The results of Mössbauer parameters (Fujita et al., 1999) (Table 3) and SEM images of the precipitates from the two sites show they have small pore diameters of about 1 nm. The surface of the precipitates from Sensui is rougher than those from Namazuta. Since this material has a low degree of crystallinity, it is not possible to give a precise crystal structure. The results from the Mössbauer effect indicate the Fe^{3+} species consists of octrahedra with a coordination number of 6.

Table 3. Mössbauer parameters of the precipitates from Namazuta and Sensui.

Sample	Δ (mm s^{-1})	ΔE (mm s^{-1})	Γ (mm s^{-1})	R.I. (%)	A.I.
Namazuta	0.35	0.75	0.50	15.78	7.92
Sensui	0.37	0.68	0.60	6.55	3.90
	After adsorption of phosphate ion				
Namazuta*	0.38	0.78	0.55	8.12	4.47
Sensui*	0.41	0.66	0.54	8.00	4.33

δ Isomer shift.
Δ Quadrupole splitting.
Γ Line width (FWHM)
R.I. Relative Intensity
A.I. Absorption Intensity

DISCUSSION

Chikuhou Coalfield, which was once a major coal producing area closed over sixty years ago, was located on the Onga River watershed in the Chikuhou region, northern part of Fukuoka Prefecture, Kyushu Island. At the present there are no active collieries in Japan.

The water from the mines and nearby springs is coloured red by a precipitate known as 'Akamizu', which is deposited on the riverbed and around the springs. The mine water generally has high electric conductivity, presumably from the dissolution of iron and sulphate ions. This water is not used in the domestic or agricultural supply but may be used in emergencies to fight large fires.

This study investigated the water quality of the mine water, the Onga River and the mineralogy and chemistry of the precipitate. Measurements of water quality include pH, electrical conductivity and sulphate concentrations, and were taken from sampling points used in previous studies.

The higher values of sulphate and pH indicate that oxidation is occurring more rapidly at Sensui than at Namazuta. It is suggested this is due to the greater activity oxidizing bacteria at Sensui as indicated by less reddening at the surface around the overflow under low pH conditions. At distance from the mine water outlet, the river becomes reddish and 'Akamizu' is deposited. Therefore, it is thought that over a long time (semi-permanently) the oxidised iron ions continues to exchange slowly, as indicated by constant values of electrical conductivity. The mine water at Namazuta is rapidly oxidised and settles more slowly. The results of suspended solids, electrical conductivity and pH indicate that the mine water contains a lot of Fe^{2+} and SO_4^{2-} and is altered by activate faecal coliforms.

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