Mine-water chemistry: the good, the bad and the ugly

David Banks · Paul L. Younger · Rolf-Tore Arnesen · Egil R. Iversen · Sheila B. Banks

Abstract Contaminative mine drainage waters have become one of the major hydrogeological and geochemical problems arising from mankind's intrusion into the geosphere. Mine drainage waters in Scandinavia and the United Kingdom are of three main types: (1) saline formation waters; (2) acidic, heavy-metal-containing, sulphate waters derived from pyrite oxidation, and (3) alkaline, hydrogen-sulphide-containing, heavy-metal-poor waters resulting from buffering reactions and/or sulphate reduction. Mine waters are not merely to be perceived as problems, they can be regarded as industrial or drinking water sources and have been used for sewage treatment, tanning and industrial metals extraction. Mine-water problems may be addressed by isolating the contaminant source, by suppressing the reactions releasing contaminants, or by active or passive water treatment. Innovative treatment techniques such as galvanic suppression, application of bactericides, neutralising or reducing agents (pulverised fly ash-based grouts, cattle manure, whey, brewers' yeast) require further research.

Key words Mine-water chemistry · Acid mine drainage · Mine-water pollution · Mine-water treatment

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Objective

The objective of this paper is to review the variety of hydrochemical characteristics, environmental impacts and treatment methods pertaining to waters discharging from mines or mine waste deposits. The paper will draw predominantly on examples from the United Kingdom and Norway, many of which have not previously been reported in mainstream literature, to illustrate this variety. A list of references is provided for the reader wishing to obtain further details of the sites reviewed.

Introduction

In the "First World", many coal and metal ore mines have been closed, for economical or political reasons, during the past decade (Younger 1994; Coldewey and Semrau 1994; King 1995; Dumpleton 1995). Such closures can lead to a variety of environmental impacts, some of the most acute being the effects on the hydrogeological environment due to cessation of pumping. Such negative hydrogeological impacts are described by Younger (1993, 1994) and may include inundation of low-lying areas (particularly areas affected by subsidence which may be regional and up to several metres magnitude; Phillips and Hellewell 1994; Yao and Reddish 1994), contaminant mobilisation and transport via rapid groundwater flow pathways (i.e. mining fractures and openings), subsidence of shallow workings and elevated mine gas emissions.
All of these problems are potentially serious, but this paper will focus on the problem known as "acid mine drainage" (although it is not always acid, nor always a problem) as it presents a particularly acute and urgent challenge to many countries (NRA 1994; King 1995). The problem is not a new one, but has been present in some guise since the commencement of deep mining necessitated the dewatering of drainage water to surface watercourses. In fact, the names of some rivers, such as the north Cornish Red River, the Ochre Dykes of Derbyshire and South Yorkshire, the Yellow Stream of Lancashire, the Norwegian Raubekken and the Spanish Rio Tinto indicate the historical nature of mining-related water contamination.

Chemical evolution of mine waters

Pyrite oxidation

Mining allows the introduction of oxygen to the deep geological environment and thus the oxidation of minerals which are in a reduced state. The same occurs when reduced minerals are brought to the surface and deposited in spoil tips (Wiggering 1993a, b). The most common family of such minerals are the sulphides. Unlike most geochemical weathering processes, the oxidation of some sulphides (of the type MS_2) leads to the production, rather than the consumption, of protons (i.e. acid). The iron disulphide, pyrite, is ubiquitous in most metal sulphide and coal deposits and may exist in potential association with other chalcophile elements such as As, Bi, Cd, Co, Cu, Ga, In, Hg, Mo, Pb, Re, Sb, Se, Sn, Te and Zn (Spears and others 1994). Pyrite undergoes a complex cycle of reactions during oxidation (Eq. 3; Fig. 1), but in simplified form the following equation describes the net processes:

$$2FeS_2 + 2H_2O + 7O_2 = 2Fe^{2+} + 4SO_4^{2-} + 4H^+_{(aq)} \quad (1)$$

Pyrite + water + oxygen = ferrous iron + sulphate + acid

Further partial oxidation of ferrous to ferric iron consumes some protons:

$$4Fe^{2+} + 4H^+_{(aq)} + O_2 = 4Fe^{3+} + 2H_2O \quad (2)$$

Ferric iron may act as an electron acceptor for further pyrite oxidation, or hydrolysis may occur, both processes releasing further protons:

$$FeS_2 + 14Fe^{3+} + 8H_2O = 15Fe^{2+} + 2SO_4^{2-} + 16H^+_{(aq)} \quad (3)$$

$$Fe^{3+} + 3H_2O = Fe(OH)_3 + 3H^+_{(aq)} \quad (4)$$

The overall sequence of reactions is acid-producing:

$$4FeS_2 + 14H_2O + 15O_2 = 4Fe(OH)_3 + 8SO_4^{2-} + 16H^+_{(aq)} \quad (5)$$

Where mine-water pumping is constant and the mine-water level is stable, little pyrite oxidation occurs below the water level, and few metals are leached from above, resulting in a relatively non-environmentally aggressive mine water. Active pyrite oxidation will, however, continue to occur in the unsaturated zone and, if pumps are turned off, the rising water level will leach out a large amount of heavy metals, resulting in a highly acid and contaminating solution. This affect has been observed in the Durham Coalfield by Younger (1993) and Younger and Sherwood (1993), as well as at Wheal Jane tin mine. Other sulphide minerals will oxidise similarly to pyrite, releasing heavy metals and sulphate. It is interesting to note, however, that oxidation of common sulphides of the form MS does not in itself release acid, e.g. sphalerite:

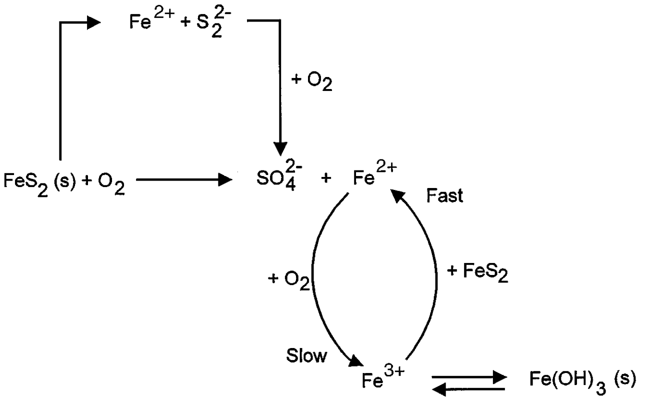

Fig. 1
Simplified diagram illustrating reaction pathways for pyrite oxidation (after Stumm and Morgan 1981; Arnesen 1993b)

$$ZnS + 2O_2 = Zn^{2+} + SO_4^{2-} \quad (6)$$

Subsequent hydroloysis of metal ions may release protons, but equilibrium speciation modelling of sphalerite oxidation-dissolution (using MINTEQA2 and/or WATEQ4F) indicates that pH is unlikely to be depressed below 5.5 due to partial hydrolysis of zinc ions.

Buffering reactions

The acidity generated by such reactions only results in a significantly depressed pH in the mine water when it exceeds (on an “equivalents” basis) the alkalinity available in the system. Such alkalinity may be available as bicarbonate in groundwater (generated by normal weathering processes or reduction processes) or in the form of mineral phases.

The most prolific mineral sources of alkalinity are the common carbonate minerals, calcite and dolomite (Nilsen and Grammeltvedt 1993). These are not, however, always present in significant quantities: in the Coal Measures of northern England, for example, the most common carbonate minerals are the ferrous varieties ankerite [$(Ca,Mg)Fe^{II}(CO_3)_2$] and siderite ($Fe^{II}CO_3$). Dissolution of these minerals will allow localised and temporary neutralisation only, for when oxidation and hydrolysis of their ferrous iron finally occurs, there will be a release of protons (see Eqs. 2 and 4), which will largely (if not completely) negate the previous neutralisation (Morrison and others 1990).

In many coal mines, limestone dust is spread liberally over mine surfaces to hinder the mobilisation of explosive coal dust and to act as a flame “barrier” in the case of an explosion. This limestone dust may also serve to neutralise mine water from working mines. This may be one of the reasons why the pumped mine water at mine 3, Longyearbyen, Svalbard (World Coal 1995) emerges at a pH of 8.2, despite a sulphur content of some 1% in the Longyear Coal (Amundsen 1994; Banks 1996; see Table 1). Additionally, it is believed that the alkaline mine water here is also due to the natural hydrochemistry of the sub-permafrost groundwater entering the mine at its deepest levels.

In the absence of readily soluble carbonate minerals, dissolution of alumino-silicate minerals (mafic silicates, feldspars and clays) can make a significant contribution to

Table 1

Comparison of discharge water quality from selected British and Svalbardian pumping and abandoned mines and spoil tips (sources; Younger 1993; Madawee 1994; Fuge and others 1991; Lemon 1991; Buchan 1962; Banks 1996; Banks and others 1996b)

	Discharge (l/s)	pH	TDS (mg/l)	Fe (mg/l)	Al (mg/l)	Mn (mg/l)	Zn (mg/l)	Cu (mg/l)	Cl^- (mg/l)	SO_4^{2-} (mg/l)
Pumping coal mines										
Kibblesworth (Durham)	740	7.1	3185	0.63			0.056			690
Nicholsons (Durham)	90	7.1	3100	5.8			0.034			1170
Kimblesworth (Durham)	105	7.3	1800	5.0			0.030			380
Tilmanstone (Kent)		7.7	2107	25					795	404
Moorgreen Piper (E. Midlands)		6.9–7.9		< 0.1–7.0					3600–10 800	
Longyearbyen Mine 3 (Svalbard)[a]	c. 0.06	8.2		< 0.01	< 0.02	0.004	0.055	< 0.005	236	7.4
Abandoned coal mines										
Stony Heap (Durham)		6.3	630	2.2						148
Dunston (Chesterfield)	c. 20	6.3		10.6	< 0.045	1.26	< 0.007	< 0.007	26	210
R. Hipper Discharge (Chesterfield)	c. 0.75	3.6		101.3	17.3	4.02	0.221	0.007	29	1044
Duke's Level, Buxton (Derbyshire)	c. 20	6.3		4.9	0.078	0.36	0.048	0.005	18	83
Ynysarwed (South Wales)	c. 35	4.2		180	< 0.5	6.1	0.061		32	1554
Hapton (Lancashire)	c. 9	7.9		0.097	< 0.01	0.138	< 0.005	0.0007	21	176
Coal spoil tips										
Crook (Durham)		3.5	1000	70						810
Quaking Houses (Durham)		4.1	2314	15						1358
Oatlands (Cumbria)		5.5		287	0.97	5.2	0.05	< 0.007		146
Thurcroft (S. Yorkshire)[b]		6.8		18.6	< 0.045	2.0	< 0.007		511	1327
Longyearbyen Mine 3 (Svalbard)	c. 0.1	3.7		1.6	1.8	0.40	0.49	0.014	4.5	77
Sverdrupbyen Mine 1 (Svalbard)	c. 0.25	2.7		179	27.5	3.2	1.3	0.168	7.0	1077
Metal mines										
Cae Coch (Wales, pyrite)		2.5		1460	84.21	3.05	0.94	0.16		5110
Cwm Rheidol (Wales – Pb, Cu, Zn)		2.8–3.0			13.9–20.1		38–72	0.03–0.068		441–846
Allen Hill Spaw (Matlock, Derbyshire)	c. 0.15 l/s	6.5		14.9	0.132	2.10	0.029		82.6	124
Metal mine spoil tips										
Cwm Rheidol (Wales – Pb, Cu, Zn)		2.6–2.7			104–128		577–978	1.2–9.35		791
EC limits for drinking water		6.2–8.5[c]		0.05[c] 0.2[d]			0.10[e]			25[c] 250[d]

[a] The alkaline nature of this water may be partially explained by the use of limestone dust to hinder explosion in the mine, but is probably dominated by the natural hydrochemistry of the deep, cold subpermafrost water entering the mine (Banks 1996)

[b] The saline water derived from this spoil tip probably represents leaching of saline pore water from the spoil derived from this deep mine. Wiggering (1993a) reports a similar phenomenon from the Ruhr area of Germany

[c] Guide level

[d] Maximum allowable concentration

[e] At discharge from pump or treatment plant

neutralisation. However, the rates of dissolution of these minerals are often significantly slower than those of the carbonates, and hence non-equilibrium rock-water interaction can be expected where flow is reasonably rapid. In particular, more unstable (mafic) silicate mineral phases such as olivine, pyroxene or anorthitic plagioclase will be more effective at neutralising acidic waters. This is illustrated by the case of Nikkel og Olivine AS's nickel sulphide mine at Ballangen, Nordland, Norway (Arnesen and Iversen 1995), where the high content of olivine around the ore body leads to a slightly alkaline mine water, with dissolved nickel and, to some extent, arsenic, being the only significant metallic contaminants. It should be emphasised, however, that simple acid-base accounting will not be able to predict mine-water quality. Relative rates of reaction and consumption of carbonate, silicate and sulphide phases will be decisive.

In particularly aggressive (low pH) mine waters, even clay minerals may undergo reaction and provide some neutralising capacity; for example, kaolinite:

$$Al_2Si_2O_5(OH)_4 + 6H^+ = 2Al^{3+} + 2H_4SiO_4 + H_2O \qquad (7)$$

This explains the high aluminium concentrations often found in mine waters of pH 4 or lower.

Sulphate reduction

In some mine systems, particularly those overflowing from saturated workings (Fig. 2), opportunities for bacterially catalysed sulphate reduction may occur, generating metal sulphides and/or hydrogen sulphide and alkalinity (Younger and Bradley 1994; Younger 1995b). The pH of the water is thus elevated. In extreme cases (for example, at Hapton Colliery in the Lancashire coalfield; Table 1), pH values of around 8 can occur, in conjunction with negligible quantities of toxic metals and considerable amounts of hydrogen sulphide. The diffusion of methane into workings may be a factor in promoting highly sulphate-reducing conditions.

Ochre deposition

The iron discharged in coal mine drainage is typically largely in the form of ferrous iron (Hedin and others 1994). Above a pH of 4 (most coal mines), ferric iron is typically absent, due to rapid hydrolysis and precipitation. Below a pH of 2.5 (e.g. in many spoil tips or sulphide mines), hydrolysis proceeds only very slowly and ferric iron can remain substantially in solution. On exposure to the atmosphere, ferrous iron will tend to be increasingly oxidised to ferric, accompanied by hydrolysis and at least partial precipitation as an iron oxyhydroxide (ochre):

$$4Fe^{2+} + O_2 + 4H^+ = 4Fe^{3+} + 2H_2O \qquad \text{oxidation (8)}$$

$$Fe^{3+} + 3H_2O = FeOOH + H_2O + 3H^+ = Fe(OH)_3 + 3H^+ \qquad \text{hydrolysis (9)}$$

Other heavy metals will co-precipitate with iron oxyhydroxide or be adsorbed onto the iron oxyhydroxide mass.

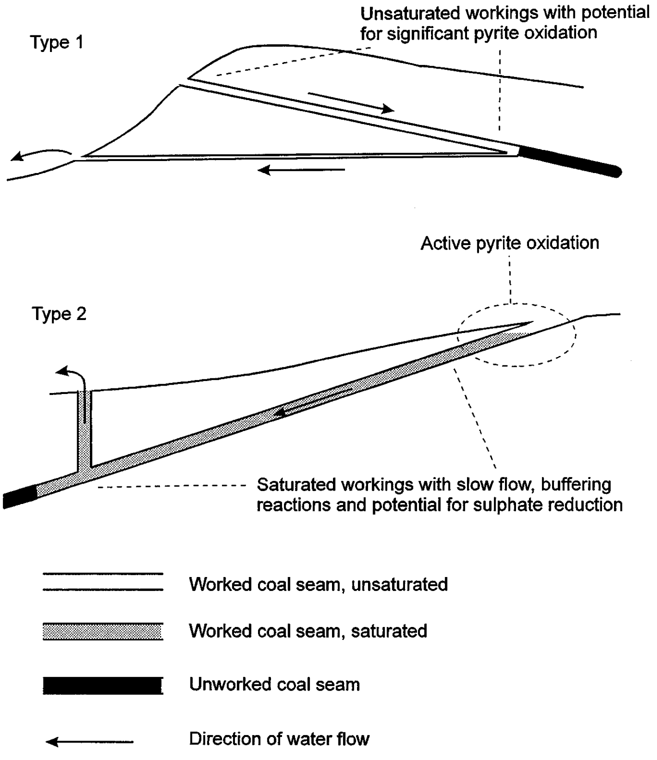

Fig. 2
Schematic illustration of two types of mine system, characterised by (Type 1) rapid unsaturated flow and intense pyrite oxidation, yielding net acidic waters and (Type 2) slow, saturated flow, buffering reactions and sulphate reduction, yielding net alkaline waters

In very low pH waters ($pH < 4$), dissolution of aluminium may occur from alumino-silicate minerals such as kaolinite. When aluminium-rich acidic mine waters are effluent into higher pH surface waters, they typically precipitate white aluminium hydroxide (gibbsite).

$$Al^{3+} + 3H_2O = Al(OH)_3 + 3H^+ \qquad (10)$$

Role of bacteria in acid mine drainage

The oxidation of pyrite in mine drainage waters is catalysed by the action of acidophilic sulphide-oxidising bacteria, in particular, *Thiobacillus ferrooxidans*, which thrives at a pH range of 1.5–3.0. *Thiobacillus ferrooxidans*, a chemoautotroph, derives energy for its metabolic processes from the oxidation of reduced sulphur and iron compounds and utilises CO_2 as a carbon source. By catalysing the oxidation of ferrous sulphide to ferric sulphate, this bacterium greatly accelerates the otherwise slow chemical oxidation of iron sulphide.

Although *Thiobacillus ferrooxidans* is probably the most well-known and predominant bacterium associated with acid mine drainage, a range of other iron/sulphur-oxidising bacteria exist which have been isolated from coal spoil heaps and drainage waters (McGinness and Johnson 1993). These include *Leptospirillum ferrooxidans*, *Thiobacillus thiooxidans* and *Sulfobacillus thermosulfidooxidans*. Spoil heaps and mine drainage waters can be expected to contain a mixed population of these and other acidophilic bacteria, the exact species present and their relative activities depending on the prevailing conditions, including temperature and pH (Norris 1990).

Bacteria are known which promote and/or catalyse the oxidation, hydrolysis and precipitation of dissolved iron (ochre formation), although their role in mine water chemistry has yet to be fully understood (McGinness and Johnson 1993; Hedin and others 1994). It is thought that abiotic oxidation of dissolved ferrous to ferric iron is fastest at elevated pH levels and, hence, in net alkaline waters with pH 6–7, this process should dominate. At lower pH values (2–3) bacterial oxidation is at its fastest and dominates in substantially acidic waters. The use of metal-immobilising bacteria has been discussed as a potential treatment method for heavy-metal contaminated effluent (Gadd 1990; Banks 1992).

Other quality problems associated with mine drainage

The oxidation of sulphide minerals to release heavy metals, sulphate and acid is the fundamental reaction characterising acid mine drainage. The quality of mine drainage water may, however, be detrimentally affected by other parameters than these, of which the most prominent are probably the following:

1. Salinity-related parameters – deep mining may encroach upon bodies of stagnant, or even fossil, groundwater of high salinity. Dewatering of coastal mines may also cause the intrusion of modern seawater. In all these cases, a high salinity may be expected in the mine drainage water, potentially resulting in contamination of fresh surface water or groundwater with chloride. This was the case at Tilmanstone colliery in the Kent Coalfield, U.K. (Headworth and others 1980), where mine drainage water containing around 318,000 tonnes of chloride at concentrations of up to 5,000 mg/l was discharged into the Chalk aquifer between 1907 and 1974. In deep saline brines in UK coalfields (Andersen 1945; Downing and Howitt 1969), elevated concentrations of ammonium in discharge water may also pose a pollution threat (NCB 1982; NRA 1995). In some deep saline mine waters, reduction of sulphate occurs, permitting the solution of high concentrations of Ba and Ra in the absence of sulphate solubility controls (Lebecka and others 1994).
2. Nitrates – where N-based explosives have been extensively used for mining or quarrying, residues of explosives in the spoil or in the mines themselves may be oxidised to nitrates and mobilised in groundwater. This problem resulted in the closure of public water supply boreholes down-gradient of a tunnel drift at Kopperå, Norway (T. Moseid, personal communication). In Ballangen mine, Norway, suspended solids, drilling mud, lubricating oil and residues from explosives are cited as significant contaminants in the mine water (Arnesen and Iversen 1995).
3. Organic parameters – the organic quality of coal mine drainage water has not been studied in detail, but trace contamination by organic compounds derived from coals is possible. One study performed in the lignitiferous regions of Romania, Serbia and Bulgaria found a tentative correlation between (as yet unidentified) refractory fluorescent organic substances in groundwater, possibly derived from contact with lignite strata, and the incidence of Balkan endemic nephrosis, a kidney complaint (Goldberg and others 1994).

The good: useful mine waters

Potable mine waters: lead mining, Derbyshire, UK

The Carboniferous limestone of Derbyshire has been mined extensively for many centuries for Pb and fluoride. The minerals fluorspar and galena, with associated sphalerite, barite, calcite, pyrite and chalcopyrite occur together in hydrothermal veins. Sub-vertical veins or “rakes” were mined at several underground levels to impressive depths. The mining often penetrated below the water table and the mines were drained by free-flowing adits or “soughs” to local rivers. It is perhaps surprising that these soughs (and also lead mining shafts) serve as high-quality, large-scale drinking water supplies today, operated by regional water undertakings (Edmunds 1971).

Table 2 demonstrates the generally wholesome nature of the water from these soughs. As might be expected for a limestone groundwater, the water is hard, rich in Ca and slightly alkaline. Sulphate levels are somewhat elevated with respect to EC guideline levels and other nearby Carboniferous lithologies such as the Millstone Grit (Edmunds and others 1989), indicating that some sulphide oxidation or sulphate dissolution is occurring. Although Zn and Ba are present at slightly higher levels than are desirable, heavy metals generally, and Pb in particular, pose no major water quality problems. There are several possible reasons why the fluoride and toxic metals are not present in excessive concentrations (Albu and others 1997):

1. Kinetic factors – the relatively slow rate of dissolution and oxidation of sulphide minerals compared with the rapid flux of limestone groundwater through the mine conduits.
2. The solubility of heavy metals is suppressed by the high alkalinity of the water and the solubility of fluorides by the high Ca concentrations. Geochemical speciation modelling indicates that Ba concentrations seem to be limited by barite solubility. Barite ($BaSO_4$) and calcite are the only two common minerals (apart from the aluminium minerals) with respect to which the waters are saturated. Minerals such as fluorite and otavite approach saturation in some waters and, for Zn and Pb, the respective carbonates yield saturation indices (SI) in the vicinity of –1. These figures may be regarded as consistent with saturation of waters with respect to fluorite and heavy metal carbonates occurring in the immediate vicinity of the ore body (and thus limiting solubility), the water later being diluted

Table 2
Water quality from lead mine soughs in the Carboniferous Limestone of Derbyshire (data from Edmunds and others 1989)

	pH	EC (μS/cm)	Ca (mg/l)	HCO_3^- (mg/l)	SO_4^{2-} (mg/l)	F (μg/l)	Ba (μg/l)	Cu (μg/l)	Cd (μg/l)	Fe (μg/l)	Pb (μg/l)	Zn (μg/l)
EC limit	6.2–8.5[a]	400[a]	100[a]		25[a] 250[b]	1500[b]	100[a]	100[c]	5[b]	50[a] 200[b]	50[d]	100[c]
Meerbrook Sough	7.2	580	86	262	57	680	70	0.9	1.6	<0.5	17	338
Hillcarr Sough	7.2	540	88	261	36	750	280	0.6	<0.5	10.2	<6	297
Mandale Sough	7.1	530	98	257	27	610	327	0.4	0.8	<0.5	16	85
Magpie Sough	7.2	520	98	261	33	640	336	0.4	0.8	<0.5	<6	74
Waterloo Inn Sough	7.0	420	98	222	30	320	229	0.5	<0.5	0.8	<6	7

[a] Guide limit
[b] Maximum permissible concentration
[c] At discharge from pump or treatment plant
[d] At point of consumption

by normal limestone groundwater during its passage along the sough.

3. The surfaces of metal sulphides being coated with insoluble oxides or carbonates, preventing further oxidation.
4. The fact that a hydraulic equilibrium situation has long been in place, with limited water-table fluctuation and thus limited flux of oxygen into the veins to promote sulphide oxidation.
5. The metals ores having been extremely efficiently removed from the veins by largely manual working.

The lead mines of Derbyshire thus provide a clear demonstration of the importance of the host rock lithology (rather than the ore mineralogy) for the quality of the mine drainage water.

Virtually identical conditions obtain in the former lead mines of the Greenhow district, North Yorkshire, where a major nineteenth century drainage adit (the Eagle Level) is exploited for public water supply by Yorkshire Water plc. Still further north, in the Pb-Zn orefield which straddles the Northumberland/Durham border, water from the abandoned Hunstanton mine was formerly used for water supply in and around the village of Blanchland. However, other mine waters in that orefield are rather less wholesome than those listed in Table 2, with major ferruginous and/or zinciferous discharges occurring in upper Weardale and South Tynedale.

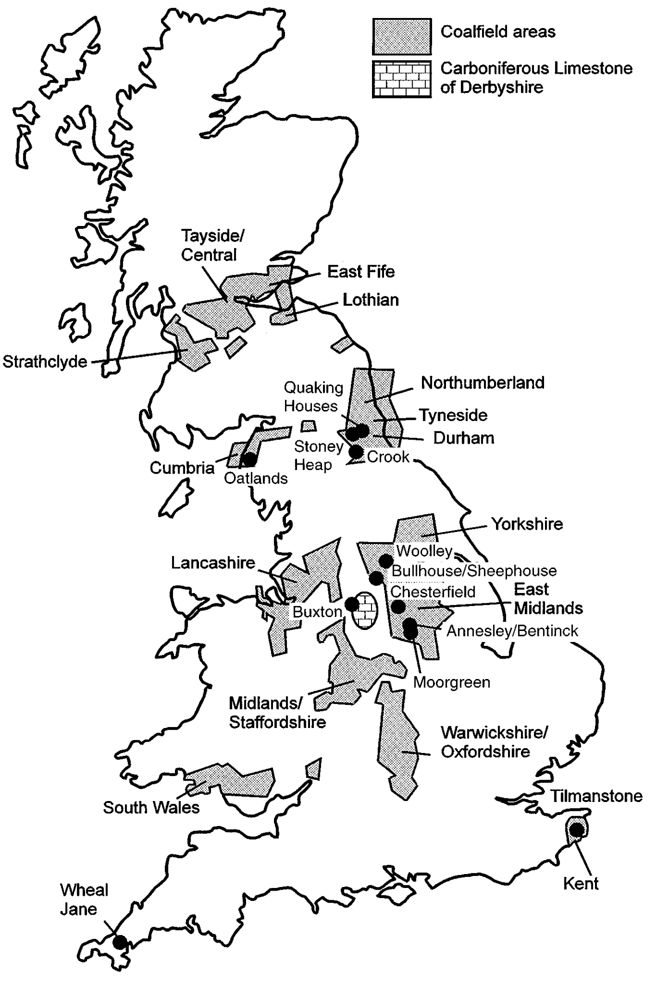

Fig. 3
Map of Great Britain, showing locations of coalfields and selected named sites

Other uses of mine waters

Banks and others (1996a) have described in some detail the other uses and positive functions of mine waters in Central and Eastern England (Fig. 3). These include:

1. The use of mine waters in sewage treatment as flocculating agents. Ochreous mine waters from the Buxton colliery in Derbyshire were, from shortly after 1886, piped some 4 km to Buxton treatment works where they were used as flocculating agents. Currently, mine water from the copper mine complex at Falun in Sweden is mixed with municipal sewage at the Främby treatment works as a form of flocculating agent, although the residue does admittedly need to be deposited at a special waste site due to content of heavy metals (some 44 mg/kg of Cd, 52.5 g/kg of Zn, 1.0 g/kg of Cu in 1990). In 1995, some 109,000 m^3/year mine water was blended with 5,900,000 m^3/year sewage (G. Strålin, Falu kommune, pers. comm.).
2. Potable water treatment – Dudeney and others (1994) proposed that ferruginous mine drainage from Bullhouse and Handbank mines near Stocksbridge, in the

South Yorkshire Coalfield, could be employed in the flocculation process for treatment of surface water from the nearby Langsett Reservoir.

3. Mine drainage waters have been employed as mineral waters or spas in historical times (Allen Hill Spaw of 1824 in Matlock, Derbyshire, UK; Naylor 1983) and are still considered for this purpose in some nations at the present day (at coal mines in China; Jianli and others 1993). Currently, it is reported that mine water (of rather good quality), derived from the Zlaté Horý gold mines, near Ostrava in the Czech Republic, is used to produce both Zlatá Vodá (Golden Water) mineral water and also carbonated soda waters.
4. In some cases, coal mine waters (particularly relatively uncontaminated pumped mine waters) have represented an important source of baseflow to otherwise overused watercourses. Such mine waters will often be free of contamination by nutrient parameters such as nitrate and phosphate, so characteristic of many effluent- or agriculture-impacted surface waters or shallow groundwaters. For example, when pumping of mine water from Strafford colliery into the sewage-impacted upper reaches of the River Dove, near Barnsley in Yorkshire, was stopped, action groups actually campaigned for the resumption of minewater pumping to support the amenity value of the watercourse.
5. In the East of England Coalfield, hypersaline brines often occur at depth as formation water in the Coal Measures strata and are encountered in deep coal mines. In the Tyneside area these were pumped industrially for their salt content. The waters formed the basis of alkali production via the LeBlanc process at Friars Goose Pumping Station from 1827 to 1927. Many of these brines were also deficient in sulphate (due to their highly reducing nature) and hence could contain large quantities of Ba. Industrial quantities of barium sulphate were extracted from pumped mine waters at Backworth Eccles colliery up until 1978.

The bad: acid mine drainage

UK examples

The British Coal Measures consist of a largely Upper Carboniferous sequence of deltaic or swamp-deposited shales, mudstones, sandstones and coals. The Coal Measures have typically been mined for coal at several levels, resulting in an "anthropogenic aquifer", a network of high transmissivity pathways stretching over long distances, sometimes across several counties, as in the East Midlands (Banks and others 1996b). Evidence suggests that, if pumped, the workings are responsible for inducing high vertical head gradients in the overlying strata (Banks and others 1997). Additionally, the impact of mankind's mining activities is seen in the widespread occurrence of ferruginous (iron-rich) mine water discharges (Younger 1993, 1995a, 1995b; Robins 1990; Banks and others 1996b).

The Great Northern Coalfield of Durham and Northumberland was once the world's most prolific source of coal. Over the last three decades, however, exploitation of the coalfield has contracted almost to zero. In the westernmost and southernmost areas of the exposed coalfield in County Durham, ferruginous mine water discharges from abandoned deep mines first emerged in the late 1970s, with marked impacts on some of the tributaries of the River Wear. Twenty years on, more than 20 discrete outfalls still emit polluted waters, typically with total iron in the range 2–40 mg/l, and with pH usually between 5 and 6.5 (Younger and Bradley 1994, Younger 1995b).

In the counties of Tyne and Wear and Northumberland, cessation of dewatering in the last decade has led to rising mine-water levels, and polluting discharges to surface waters are anticipated early in the next century (Younger 1995c). There are as yet no contingency plans for combatting this pollution and consequently serious degradation of river quality is anticipated in the Tyne, Blyth and Wansbeck catchments. The peculiar legal situation prevailing until very recently in Britain, whereby mine-water discharges from abandoned mines were specifically exempted under the Water Resources Act of 1991 from pollution legislation, has left a regulatory vacuum in which no specific party has accepted full responsibility for tackling mine-water-related problems.

Copper and zinc mining, Norway

The Caledonian mountain chain and, in particular, the base metal mineralisation province of Trøndelag (mid-Norway) hosts Norway's richest strata-bound sulphide bodies of Fe, Cu, S and Zn, and also some of Europe's worst mine drainage-related pollution (Iversen and Johanessen 1984; Fig. 4). The region also boasts some of the country's finest salmon fishing rivers and the two activities have not been able to avoid a degree of conflict. Indeed, the Orkla, Gaula, Orva, Hitter, upper Glomma, Stallvika, Skorovatn/Grøndal and Bleikvass Rivers have all been classified by the State Pollution Prevention Agency as being "significantly" or "highly" polluted (grade 3 or 4) as a result of mining activities releasing Cu, Zn, Fe, Pb and Cd (Schartau 1992).

The metals are derived from natural exposures of sulphide minerals, mine drainage, tailings, spoil and slag deposits, and processing works. In areas where mining has a long history, such as the Røros area (commercial mining for copper since 1644), the main source of pollution is usually subaerial mine-waste tips, with a high content of sulphide minerals due to inefficient extraction practices. Later, towards the end of the 1960s, the practice of underwater disposal of mining tailings from selective flotation became widespread (Arnesen and Iversen 1993), with the aim of limiting access of oxygen to the waste. Seven mines have practised underwater disposal of tailings in the recent past; at Folldal (Hjerkinn), Grong, Løkken, Røros, Skorovas and Sulitjelma, with sulphide contents in the tailings ranging from 4% to 45%.

The mines have typically been worked historically (some since the seventeenth or eighteenth century) for Cu or

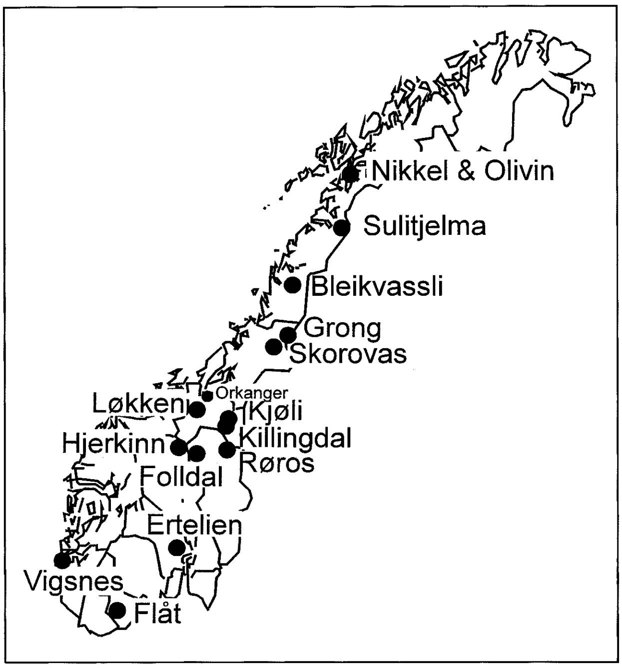

Fig. 4
Map of Norway, showing locations of selected Norwegian metal sulphide mines

Zn. More recently, FeS_2 (for sulphuric acid production) has been the economic *primus motor* in many mines, particularly in the late nineteenth and early twentieth centuries, although falls in S prices in the 1970s caused a partial reversion to metal production. Many of the mines have become uneconomical over the last two decades and, despite determined government subsidy to sustain employment, have been gradually closed down.

Skorovas mine

The Skorovas mine commenced activity in 1952/1953 with the ore containing initially 3–4% Cu. Later the composition of the ore stabilised at 45% S, 1.3% Cu, 1.7% Zn and 0.05% As (Arnesen and Iversen 1995). The sulphide ore was initially worked for S, but a fall in S prices led to Cu and Zn being the primary products since 1976. The mine closed in 1984, and the environmental consequences of Skorovas's mining history are concisely summarised by Kopperstad (1992). Up until November 1975, mining waste from ore separation was deposited in and around the shores of Dausjø Lake, whereas, after this date, underwater disposal in the lake of tailings from selective flotation took place. In total, some 200,000 tonnes of waste were produced, containing around 30% S (Arnesen and Iversen 1995).

The mine area drains to two catchments. The mine itself drains into the Stallvik River catchment, whereas the run-off from the mine waste tips and the natural outcrop of the ore drains via the Dausjøbekk Stream and Dausjø Lake into the Skorovas and Grøndal Rivers. The former catchment has received some 8–10 tonnes Cu and 17–18 tonnes Zn per year, while the latter has received slightly more. Of the metal run-off to the Skorovas catchment, some 10% is thought to be due to natural weathering of the ore outcrop. Indeed, the name Dausjø (Dead Lake) is perhaps indicative of long-standing natural "contamination" in the Skorovas catchment. Mining-induced contamination is thought responsible for the lower Stallvik River being empty of fish. The mining history of the area has left a spectrum of contaminant sources for today's environmental geologists to tackle, including the mine itself, the mine waste tips and the contaminated deposits lying at the bottom of Dausjø Lake.

Killingdal mine

The two ores at Killingdal, the North and South Ores, were found in 1674 and 1791, respectively (Table 3). The genesis and mineralogy of the ores are described by Rui (1973). Working was only sporadic prior to 1850. The mine's production was never large; by 1984, 3 million tonnes of ore had been extracted, but the mine was closed in 1986. The mine is over 1 km deep, consisting of two shafts, the Killingdal shaft (along the ore body) and the Bjørgan shaft (a transport shaft constructed, in 1965, at right angles to the ore). Prior to the 1980s, highly Cu, Zn, Fe and SO_4^{2-} contaminated run-off from spoil heaps at the top of the Killingdal shaft was allowed to flow into streams draining into the Gaula River (Iversen and Johannessen 1984). The mine was relatively dry, being constructed in low-permeability phyllites, and no deep pumping was necessary during working. The small amounts of seepage were removed partly by shallow pumping, while a certain amount (some authorities suggest 6 l/min, although the real figure is likely to be less) was removed by evaporation caused by elevated temperatures and extremely powerful air currents through the mine Tentative water fluxes prior to closure are shown in Fig. 5. Following closure of the mine, the spoil heaps were, in 1991, collected around the top of the Killingdal shaft and covered by low-permeability till. This was capped with coarse stones to hinder erosion. Drainage water from the tips was collected and diverted into the Killingdal shaft. It was anticipated that the amount of water draining from the shaft to the mine would be sufficiently small to be evaporated away within the mine system (Arnesen and Iversen 1995). Unfortunately, due to errors in estimating the mine's water balance, the till capping did not exclude sufficient water from the shaft area and water was found to be running into the mine at a rate of 40 l/min (Fig. 5).

The rate at which the mine was filling up was only uncovered when the Norwegian government authorities decided that a deep mine like Killingdal would be ideal for disposal of low- and intermediate-level radioactive waste. The team chosen to assess this proposal visited the mine to report that the mine was filling with water with a pH of 2.5, over 10,000 mg/l SO_4^{2-} and an impressive array of heavy metals, at a rate of some 13 m per month (NOTEBY 1992). The quality of the water draining down the Kil-

Table 3
Composition of Norwegian Cu/Zn sulphide ores

	Killingdal (North Ore)[a]	Killingdal (Main Ore)[a]	Skorovas (late production)[b]	Løkken Ore[c]
S (%)	42.14	48.15	45	41.4
Fe	34.15	40.99		37.5
SiO_2				13.7
Cu	1.03	1.90	1.3	2.2
Zn	10.61	5.89	1.7	1.9
Mn				0.07
Co				0.07
Pb	0.40			0.02
Ni ppm (g/tonne)				80
Se				50
As	1200		500	40
Cd				10
Ag				16
Au				0.2

[a] after Rui (1973)
[b] after Arnesen and Iversen (1995)
[c] after Arnesen and others (1994)

lingdal shaft was seen to decline with increasing depth in the mine (Banks 1993, 1994), due to either (or both) further oxidation of ore minerals in the mine or concentration increase due to evaporation (Fig. 6). By June 1994, the Central Station level was submerged and air circulation (and hence evaporation) within the mine was curtailed (Arnesen and Iversen 1995). Overflow at a rate of some 40 l/min from the Bjørgan shaft was predicted within 40 years. Even if the drainage water were to be diverted away from the Killingdal shaft, overflow would occur within some 400 years, though this rate of overflow would be unlikely to represent a major problem. The plans for radioactive waste disposal appear to have been dropped (Henriksen and others 1992), but mine water continues to accumulate, and a new solution to the runoff from the spoil tips at the mouth of this shaft may eventually have to be found.

Løkken mine, Norway

The Løkken area consists of a complex of interconnected mines in a westwards dipping FeS_2–$CuFeS_2$–ZnS ore body (Table 3) in greenstone bedrock. The mines are of varying age, active from 1664 to 1987 (Fig. 7) and are all now closed. The Løkken mine system is some 4 km long, consisting of four main shafts, the deepest being some 1,000 m deep. Until 1952, the highly contaminated, pumped mine water was discharged to the Raubekken (Red Stream), a tributary of the River Orkla. This had a major impact on salmon fishing in the Orkla and from 1952, the mine water was piped 30 km to the town of Orkanger, at the mouth of the Orkla in Orklafjord, where the water was chemically treated at the Thamshavn smelter. After 1962, however, treatment ceased and the raw mine water was discharged to the fjord. The deep Astrup shaft was opened in 1972. The water quantity

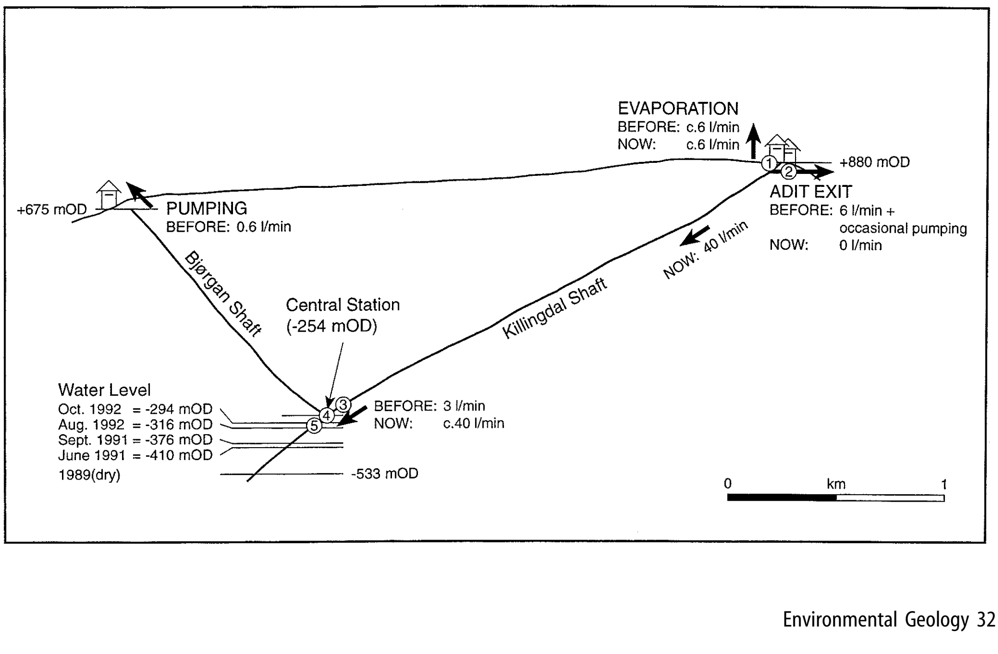

Fig. 5
A cross section through Killingdal mine, illustrating water fluxes before and after remedial action on spoil tips in 1991 (after Banks 1994). The points marked 1–5 represent the sampling locations on which Fig. 6 is based (mOD = metres above sea level)

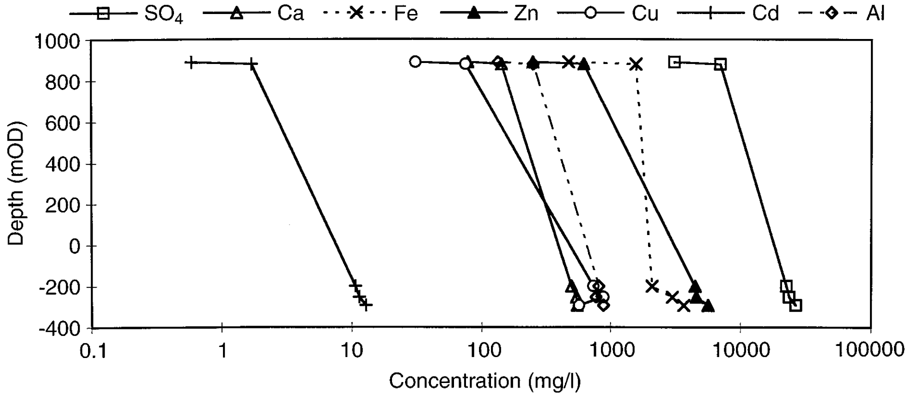

Fig. 6
Variation with depth of hydrochemistry of free draining water running into mine along Killingdal shaft (after Banks 1994). Samples taken Autumn 1992; for sampling locations, see Fig. 5 (mOD = metres above sea level)

from this part of the mine was pumped into the Wallenberg complex but comprised only some 5–10% of the total quantity pumped from the latter (Arnesen and Iversen 1995). Up until 1983, the water pumped from the mine complex was the largest single emission of Cu and Zn in Norway. The water contained 0.5–1 g/l Cu and a similar quantity of Zn, and was discharged to Orklafjord at some 500,000–600,000 m^3/year. In 1983, the old part of the mine around Wallenberg was closed and sealed off from the deeper Astrup complex with concrete plugs. The Wallenberg complex was allowed to fill up and water from the Astrup complex was also pumped at some 55,000 m^3/year into the Wallenberg mine until 1987, when the Astrup mine was closed and also started to fill up.

In 1989, when the Wallenberg mine was still filling, the major source of mining-related pollution was assessed by the Norwegian Institute for Water Research (NIVA) as being the numerous old spoil tips along the Løkken valley sides, which were responsible for 90% of the total Cu run-off. In 1992, the Wallenberg shaft had filled and pumping had to be recommenced. A solution to run-off from both the spoil tips and the mine itself was thus required (Arnesen 1993a).

The ugly: extreme visual impacts

Wheal Jane, Cornwall, UK

Wheal Jane, between Redruth and Truro, was one of the more modern Cornish tin mines, having only been extensively worked since the 1960s. However, recent workings intersect older stopes and roadways dating back hundreds (if not thousands) of years. The host rocks are

Table 4
Composition of a variety of mine waters from Norwegian sulphide mines

Mine Main ore	Ballangen[a] (Ni/Olivine)	Kjøli[b] (Cu/Zn)	Løkken[c] (Cu/Zn)	Løkken[d], Astrup Shaft (Cu/Zn)	Stowartz, Røros[e] (Cu/Zn)	Skorovas[f] (Cu/Zn)	Flåt, Evje[g] (Ni/Cu)	Killingdal[h] (Cu/Zn)
pH	7.22	2.72	2.3	2.71	6.28	2.36	3.68	2.7
Cond. (μS/cm)	457	2158			445	5922	267	
SO_4^{2-} (mg/l)	93	1564		1671		7291	68	26500
Fe (mg/l)		248	1529	210	5.06	2284		3680
Al (mg/l)				20.1			2.58	885
Cu (μg/l)	5.9	25500	530000	35600	200	129400	776	574000
Zn (μg/l)		1080	870000	131000	3300	256500	151	5640000
Ni (μg/l)	441						2150	
Cd (μg/l)				404	100	519		12800
As (μg/l)	192							280

[a] Mine water after sedimentation basin, average 1993, Arnesen and Iversen (1995)
[b] Collected run-off from Kjøli mine, average for 1984, Arnesen and Iversen (1995)
[c] Mine water, average for period 1961–1975, Arnesen and Iversen (1995)
[d] Average for water samples from the Astrup shaft, March 1988–October 1989, Arnesen and others (1994); Arnesen and Iversen (1995)
[e] Mine water, average for 1990/1991, Arnesen and Iversen (1995)
[f] Mine water from Gråbergstollen, average for 1990, Arnesen and Iversen (1995)
[g] Mine water from Gruvebekken, Arnesen and Iversen (1995)
[h] Mine water from base of Killingdal mine, after Noteby (1992) and Banks (1994)

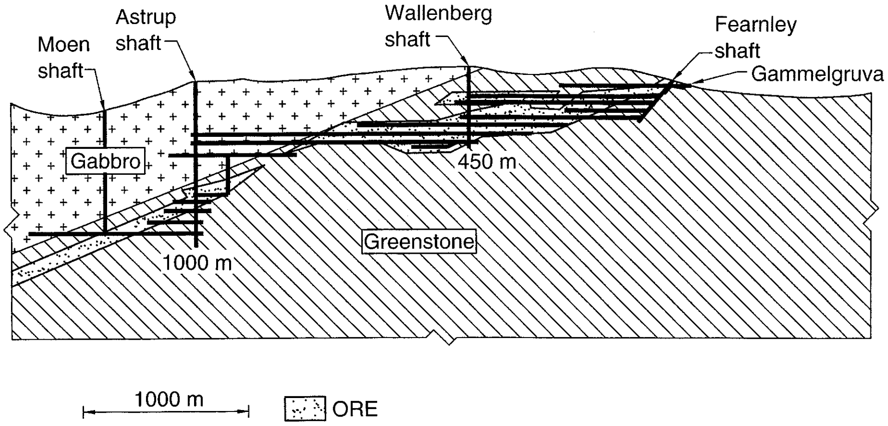

Fig. 7
Schematic section through Løkken mine, Norway

Killas shales intruded by quartz porphyry dykes which carry veins bearing cassiterite, chalcopyrite, pyrite, wolframite, arsenopyrite, galena and silver. While the tin minerals were always the most important ores, some of the more extensive old workings at Wheal Jane were among the most prolific producers of pyrite (as a by-product) in Cornwall. Additionally, arsenopyrite was worked for arsenic from the Nangiles mine (also connected to Jane). The modern workings reach depths of 450 m and were exceptionally wet by UK standards, with routine dewatering around 10^7 l/day, and peak pumping rates up to six times this amount. Recent data strongly suggest that these high flow rates arise from the huge extent of the associated old workings, rather than from any unusual permeability in the Killas host rocks (Bowen and others 1994). During mining, pumped water was discharged to the River Carnon after partial treatment. In March 1991, a UK government grant to support dewatering was withdrawn, resulting in an immediate cessation of pumping and consequent closure of the mine (Hamilton and others 1994a).

The subsequent mine water rise was monitored, both in terms of level and quality. On 5 July 1991, the water quality at three different depths was recorded (Table 5), revealing incipient stratification of the water in the mine, with the poorer water quality occurring at depth. Water levels in the mine rose extremely quickly (Fig. 8), with a recovery of more than 400 m in less than 8 months. On 17 November 1991, the mine water overflowed into the River Carnon via Jane's adit at 14.5 m above sea level. The owners had provided a treatment lagoon for the overflow, which soon proved to be embarrassingly inadequate. The initial discharge was of water with a pH of 2.8 and a total metals loading of over 5 g/l. The National Rivers Authority took emergency action by lime dosing the water in Jane's adit, plugging the adit mouth and pumping water from the adit to the Wheal Jane tailings dam. In late December 1991, pumps failed and arrangements began to be made for a more sustainable pump-and-treat operation at the No. 2 shaft. Before the new operations could commence, however, a further rise of head in the mine system of around 4 m led to a catastrophic release of an estimated 50×10^6 l of highly polluted mine water when a plug in the portal of the old Nangiles adit unexpectedly burst.

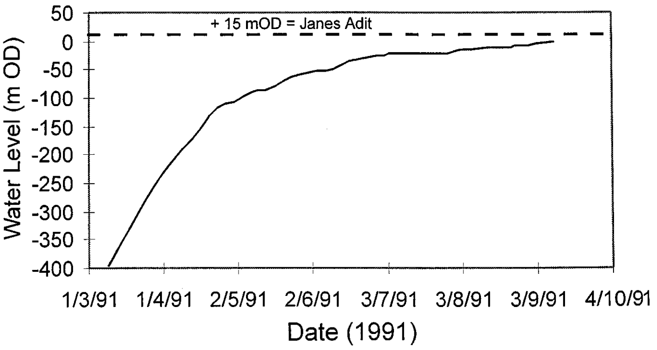

Fig. 8
Rate of rise of water levels (mOD = above sea level) in Wheal Jane tin mine, shaft No. 2, following pump shutdown in March 1991 (reproduced with permission from J. Wright, Environment Agency, UK)

Table 5
Water quality in Wheal Jane Mine, Cornwall, following refilling of mine (after Bowen and others 1994)

Sample depth	pH	Cond.	Cd	Fe	Zn	Cu
(m below water level)		(μS/cm)	(mg/l)	(mg/l)	(mg/l)	(mg/l)
Shallow 10 m	3.7	1640	0.13	94	98	4.5
Middle 90 m	2.6	7390	2.6	1846	1379	40
Deep 180 m	2.5	8440	4.8	2162	1541	44

The main consequence of the Nangiles plug failure was rapid and spectacular discoloration of the Fal Estuary. In full daylight, and beneath the cameras of national news helicopters, an orange plume spread throughout the estuarine waters, past bayfront residences, marinas and hotels. It was certainly the single most spectacular pollution incident in UK living memory. The potential damage that a repeat of such unsightly pollution could do to the local tourist industry caused justifiable alarm to residents of the area, and the national news coverage ensured that the UK government was quick to authorise emergency pollution prevention activities by the National Rivers Authority (Hamilton and others 1994b). Accordingly, pumping was initiated from the Wheal Jane No. 2 shaft, and active treatment commenced (aeration, lime dosing, flocculation and settlement in the tailings dam). The active treatment system was rapidly optimised, and now has a typical metals removal efficiency in excess of 98%. In the following 4 years of sustained pumping and treatment, discharge rates have varied between 5,000 and 40,000 m^3/day, while Fe contents in the pumped water have fallen steadily from some 1,600 mg/l to 400 mg/l, indicating an approach to a dynamic equilibrium in the system, as gradual flushing of the products of pyrite oxidation (which were largely taken into solution during groundwater rebound) reaches completion.

Biological surveys of the Fal Estuary after the initial mine-water outburst surprisingly revealed little or no acute impact on marine flora and fauna. This was especially good news, given that the Fal area contains one of only two Maerl colonies in the United Kingdom (an internationally important marine habitat, resembling coral reefs, with exceptionally rich biodiversity). It thus transpires that the principal impact of the extreme pollution from Wheal Jane was visual – the ultimate in ugly mine waters.

Living with the bad and the ugly – treatment of mine drainage

Several options exist to ameliorate the effects of acid mine drainage from disused workings:

1. Continue pumping in order to retain control over drainage pathways. This may eventually evolve into a policy of controlled refilling of mines (e.g. Durham Coalfield, U.K.).
2. Plug drainage adits and allow the mine to refill (e.g. Skorovas, Norway).
3. Treat drainage water (e.g. Wheal Jane, U.K.).
4. Dispose of drainage water to a lower sensitivity recipient: for example, seasonal storage of mine water for later release to a recipient during periods of high flow (e.g. spring flood).

Similarly, a number of options exist to minimise the contaminated run-off from spoil tips:

1. Isolate waste tips in situ by, for example, low-permeability membranes (e.g. Kjøli, Norway).
2. Move waste tips to a more suitable place of disposal, for example, underwater (e.g. Skorovas, Norway).
3. Recirculate contaminated water through mine workings (e.g. Løkken, Norway).
4. Collect contaminated water for treatment or re-infiltration.

Conventional active treatment

As the Wheal Jane case study illustrates, conventional active treatment by alkaline dosing, aeration, flocculation and settlement can provide an efficient response to mine water pollution (Best and Aikman 1983). In most treatment plants for sulphide mine drainage in Canada and Sweden, calcium hydroxide is the preferred alkali for dosing (Arnesen 1993b). Such treatment processes are aerobic, with heavy metals typically being precipitated as hydroxides.

Alternative active treatment strategies employing anaerobic processes have also been successful, as at Laisvall in Sweden, where lead is precipitated as sulphide or at Thamshavn in Norway where mine waters from the Løkken mine were treated with hydrogen sulphide produced at the Thamshavn smelting works, with copper being precipitated as a sulphide (Arnesen 1993b).

Other treatment methods which have been used for removal of heavy metals from non-ferruginous waters include the addition of scrap iron to galvanically reduce and precipitate copper from solution, ion-exchange techniques, electrochemically assisted sorption onto dead biomass (Hancock and others 1995), reverse osmosis (Arnesen 1993b) and electrolysis to recover copper (under trial at Kongens mine, Røros, Norway).

Passive mine-water treatment

Active treatment may be a cost-effective long-term solution to mine-water pollution at sites such as Wheal Jane where loadings of metals and acidity are especially high. However, where discharges from abandoned mines are less extreme in their quality, the ongoing revenue costs of active treatment can become less attractive, and passive treatment (primarily using constructed wetlands technology) is increasingly favoured (Hedin and others 1994).

In the United States, acid drainage from abandoned metals mines poses such acute problems in some areas that several localities have been accorded "Superfund Status" by the federal government (King 1995). One such area is Colorado, where gold, silver and other metals have been mined since 1800. The mines are now largely abandoned. Excess concentrations of Cu, Zn, Cd, Pb and As affect some 13,000 miles of the USA's rivers, 1,400 miles of these in Colorado alone (CDM unpub. report). One Superfund site, the Clear Creek/Central City site, lying some 35 km west of Denver, covers some 400 square miles and has been the subject of intense research on mine water treatment. Methods assessed included conventional active treatment, but the study concluded that "natural" or "passive" treatment of water was the only technically and economically feasible long-term alternative. In a joint project between Camp Dresser and McKee (CDM) and the Colorado School of Mines (CDM unpub. report), a pilot-scale treatment plant, based on reed bed technology was built and tested. The treatment plant consisted of a 3-m × 20-m × 1.3-m deep concrete trough, divided into three compartments, containing a layer of river rock, a layer of compost material or substrate planted with locally transplanted rushes and cattails. In later versions, baffles were added to increase contact with the substrate and finally an aerobic "polishing" cell was added. Under optimum conditions, the plant operated very successfully, removing up to 99% of Cu, 98% of Zn, 94% of Pb and 86% of Fe, while increasing the pH from 3.0 to 6.5.

Numerous examples of the application of constructed wetlands to drainage from abandoned coal mines in Appalachia have been reviewed by Hedin and others (1994), who conclude that predictable chemical reactions are responsible for the observed improvements in water quality, and that the most frequent cause of failure of constructed wetlands is under-sizing.

In the United Kingdom, passive treatment of mine waters has been initiated only since 1994, with the construction of an experimental wetland system at Wheal Jane, a series of wetlands in the Pelenna Valley of South Wales and at Woolley Colliery in West Yorkshire (Coal Authority/IMC, unpub. report), and a compost wetland for treatment of acidic, Al-rich colliery spoil drainage at Quaking Houses, County Durham (Younger 1995c). The long-term viability of such passive treatment systems does not, however, at present, seem to have been adequately addressed.

Other control/remediation techniques

Continued pumping

In some large, interconnected coalfields, the most cost-effective means of dealing with mine water pollution may be to continue regional dewatering after mining has ceased (Younger and Harbourne 1995). In Durham, mine-water levels in the extensive coal mine complex underlying the county are being held at bay by continued dewatering. Thus, the breakout of mine waters to surface recipients is being prevented, at least as long as central government continues to provide funding for this activity to the Coal Authority. Cessation of dewatering in this area would probably result in major mine-water pollution in the River Wear catchment. Elevated Fe levels would probably lead to either the closure or expensive major upgrade of the 26×10^6 l/day public water supply works at Lumley, based on the River Wear. In West Yorkshire, the Coal Authority operate a similar scheme at the former Woolley Colliery (Coal Authority/IMC, unpub. report). Pumping at this shaft (with associated passive settlement and wetland treatment) ensures that uncontrolled surface discharges of highly ferruginous mine water will not occur, and also prevents flooding of underground workings at the National Museum of Mining at Capstone. A further scheme along similar lines is planned for the Dysart-Leven Coalfield, Fife, Scotland (Younger and others 1995), where currently rising mine waters are being monitored until they reach a pre-set threshold some tens of metres below sea level. When this level is reached, a pump-and-treat scheme will be initiated to prevent polluting discharges in the valleys of the Rivers Ore and Leven.

Isolation – Kjøli mine, Norway

Kjøli mine is another Cu and Zn mine in the vicinity of Killingdal which contributed heavily to pollution in the River Gaula. One million tonnes of ore were mined between its opening in 1766 and its abandonment in 1941. Some 200,000 tonnes (or 80,000 m^3) of spoil have been dumped in tips around the mine area and these have been responsible for a yearly run-off of 4.2 tonnes Cu, 0.24 tonnes Zn, 38.3 tonnes Fe and 198 tonnes sulphate (Østmoe 1991; Iversen 1992). In 1981, the tips were collected in one area and were limed with 100 tonnes of calcium hydroxide (the quantity being that calculated to neutralize the run-off for 1 year). This failed to provide any long-term improvement in run-off. Indeed, following liming the situation appeared to deteriorate (Fig. 9). This is likely to have been due to the physical disturbance and hence oxygenation of the spoil tips. After a careful assessment, isolation of the spoil heaps (to hinder access by oxygen and water) was found to be the most promising course of remedial action. The isolation, which took place in 1989, employed spoil tip capping using synthetic geomembranes overlain by a natural till cover. The option of using till alone was not considered to be sufficiently impermeable to hinder access of water. Also in 1989, the mine adit was plugged by concrete. The water level within the mine rose some 17 m but, by 1990, a new overflow had occurred at a higher level (Arnesen and Iversen 1995). Nevertheless, the remedial action appears to have been largely successful. Although the yearly transport of Cu and Zn from the mine area is still around 600 kg/year Cu and 90 kg/year Zn (1992 values; Arnesen and Iversen 1995), these still represent a significant decline in Cu and Zn compared with 1985.

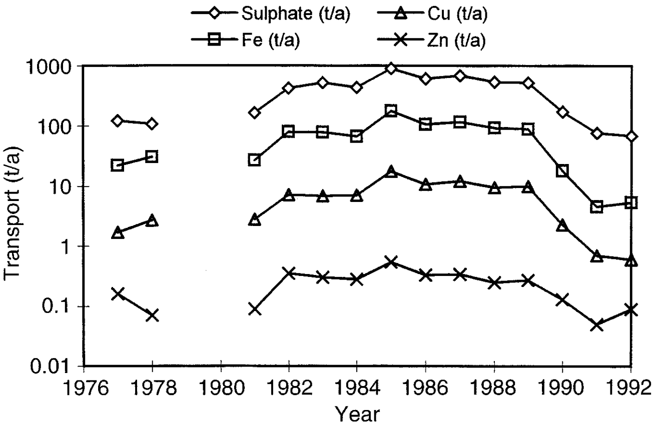

Fig. 9
Time weighted annual transport of sulphate, copper, zinc and iron (t/a = tonnes/year) from the Kjøli mine area (after data from Arnesen and Iversen 1995)

Recirculation – Løkken mine, Norway

In 1989, NIVA assessed the major part of the Cu contamination at Løkken as being generated by the numerous spoil heaps. The immediate recipient of the contamination is the highly polluted Raubekk Stream and, ultimately, the Orkla River. Several forms of action have been taken to reduce the release of heavy metals to the environment, but the most novel has been the full-scale attempt at recirculating drainage water from the spoil tips through the now water-filled mine complex (Kommunal Teknikk 1994; Lundberg 1994), using the 6×10^6 m^3 of mine space as a giant copper adsorption basin.

NIVA carefully monitored the quality of mine water in the Wallenberg mine during its filling (Fig. 10). The water developed a clear stratification, with relatively uncontaminated "surface water", derived from run-off through mine openings and open fractures, lying over deeper highly contaminated mine water. Although sulphate levels were as high as they had ever been in the deep mine water, pH had increased relative to the original mine water, Cd had decreased and the concentrations of Cu had reduced by up to 99.9%, to less than 1 mg/l in some cases. Zinc, conversely, showed elevated concentrations, of up to 4 g/l at the greatest depths (Arnesen and others 1994; Arnesen and Iversen 1995). The conditions did not appear suitable for sulphate reduction, and it was thus suggested that Cu adsorption and metal exchange for Zn was taking place on the surface of sulphide minerals, and that this could be used to treat spoil tip waters by circulation through the mine. As Zn is considered to be a lesser environmental toxin than Cu, the overall effect is an improvement in quality.

The treatment system commenced between April and August 1992. Spoil tip drainage is introduced at a rate of 600,000 m^3/year to the mine complex at the tourist mine Gamlegruva (Old Mine) and, after a journey of approximately 1.2 km, taking several years, is pumped up from the newer Wallenberg shaft (Kommunal Teknikk 1994). The workings contain a considerable quantity of basic lithologies, such as greenstone and calcite, which contribute towards raising the water's pH. The other parameter most radically affected is Cu. Whereas the original water contains 75–200 mg/l Cu, the pumped effluent water contains a mere 0.5–10 mg/l Cu and around 20 mg/l Zn (Arnesen and others 1994; Arnesen and Iversen 1995). It is perhaps a little too early to judge the results, however, as it is not certain that full breakthrough of influent water has been achieved in the Wallenberg shaft. There are also some doubts as to the long-term sustainability of the effect, as exposed pyrite is eventually expected to become saturated with Cu, but the process is imaginative, promising and deserves further research.

Longer term plans may involve attempts to promote sulphate reduction, and hence metal sulphide precipitation, within the mine. This might be achieved by adding an organic substrate to promote the growth of sulphate-reducing bacteria. Laboratory-scale research is being carried out on the use of surplus dairy whey, cow manure and brewing yeast as "food" for bacterial communities (Lundberg 1994; E. Iversen, pers. comm.; Arnesen and others 1994).

Hydraulic control – Killingdal mine, Norway

One option which is currently being discussed (Henriksen and others 1992) is the possibility of "plugging" the Bjørgan shaft with a concrete block, thus preventing mine water overflow. In order for this to work, however, several conditions must be satisfied:

1. No additional adits should exist. In older mines, accurate plans may not exist and it may not be possible to locate all old adits and workings.
2. The plug must be engineered to withstand the 200 m head difference between the Bjørgan and Killingdal shafts and be constructed of a material capable of withstanding the potentially acid and sulphate-rich nature of the drainage.
3. The rock should be of sufficiently low permeability that mine water may not penetrate natural groundwater flow paths (e.g. fractures) to emerge as contaminated springs.
4. The question of how to deal with the continuing production of acid drainage by the spoil heaps must be tackled.

Alternatively, the spoil tips could be re-capped to minimise the amount of leachate entering the mine. The reduced flux of water could be allowed to flow through the workings to exit at a low rate from the Bjørgan shaft. It might be expected that the quality of any water which is

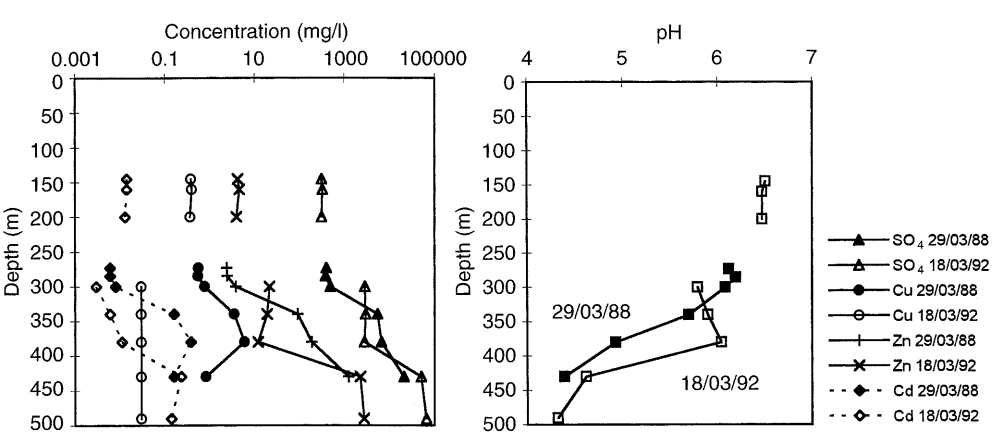

Fig. 10
Variation in water chemistry with depth in the water-filled Wallenburg shaft, Løkken, during filling of the mine (after data from Arnesen 1993b)

allowed to exit from the Bjørgan shaft will be superior to that entering the mine, due to exchange or precipitation of heavy metals taking place at depth in the mine (see section on Recirculation – Løkken above). A first flush of highly polluting water may yield problems.

Combined solutions – Skorovas mine, Norway

Improvements to the Dausjø catchment commenced in 1976, with the underwater disposal of tailings from selective flotation. Lime is often incorporated in the flotation procedure and tailings have an elevated pH (at least temporarily until pyrite oxidation exhausts the buffering capacity of the lime). The impact of the disposal of the alkaline tailings on heavy metals and pH in Dausjøbekk is seen in Fig. 11. This has been responsible for re-establishing fish life in the Grøndal River.

As discussed above, the Skorovas mining area contains several sources of contamination, all of which have had to be dealt with in a clean-up programme (Kopperstad 1992). Concerted environmental rehabilitation began in 1984 when the contaminated deposits lying in Dausjø Lake were capped with relatively inert ground rock waste. Acid mine drainage from spoil heaps was diverted past Dausjø to encourage a rise in pH and thus hinder mobilisation of metals from the contaminated sediments. In addition, a liming station was established on Dausjø Stream to raise the pH of the water.

These remedial works were only partially successful. To control contamination in the watercourse, 400 tonnes of lime per year were required, while the metal hydroxides precipitating out downstream of the liming station were causing a significant problem in themselves. In 1987–1988, it was therefore decided to negate the need for liming by removing the major source of the acid drainage contamination to the Skorovas catchment, namely the spoil tips. The favoured location for their removal was the bed of the 20 m deep Dausjø Lake. The lake was partially emptied and the spoil emplaced and covered with till material. The lake was then refilled, and it was hoped that metals would be immobilised in the low-throughflow and probably chemically reducing environment of the lake bed. The lake was refilled in 1991 and results so far appear promising. Heavy metals in the catchment are being held at satisfactory levels without the need for liming (Kopperstad 1992; Arnesen 1993b).

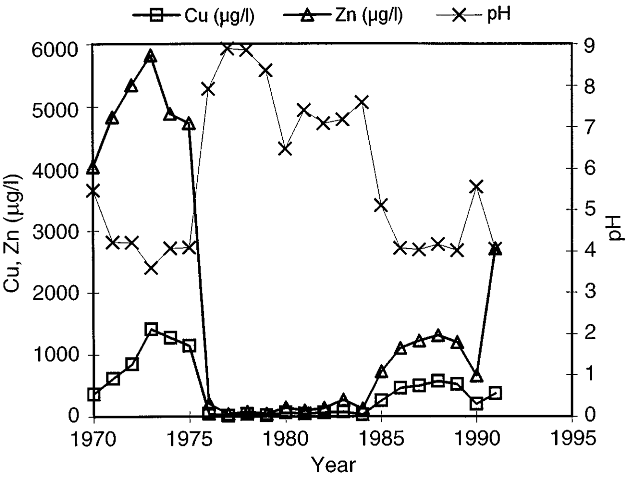

Fig. 11
Water quality at the exit from Dausjø Lake, Skorovas mine (after data from Arnesen 1993b)

As regards the mine itself, the chosen solution was to seal off, in 1990, all the lower adits and passageways with concrete plugs and allow the mine to refill. Currently, now that the mine drainage no longer discharges, improvements are noted in river water quality and fish have returned to Stallvik River. The mine is expected to fill up and overflow from an upper adit in 1995–1997. It is anticipated that reducing conditions will be reestablished at the deeper levels of the mine, hindering further sulphide oxidation and that the rate of throughflow will be small compared with the original active mining condition. It is thus anticipated that, although some continued discharge of acid mine drainage from the mine will occur, the refilling of the mine will represent a substantial environmental improvement.

Innovative technologies

Several authors have proposed the use of imaginative techniques, which probably require considerable further research before they can be applied reliably "in-mine". Shelp and others (1995) have shown at laboratory scale that the use of sacrificial anodes can suppress the oxidation of metal sulphide ores. It is also speculated whether, in nations which are energy-rich, such as Norway, a potential difference could be applied to the ore body, opposing the natural galvanic couple generated by ore oxidation (Logn and Bølviken 1974; Bølviken and Logn 1975) and thus suppressing the oxidation reaction.

The oxidation of sulphides is typically bacterially catalysed. Rastogi (1994) has demonstrated that the application of bactericides can hinder pyrite oxidation in spoil tips. The application of bactericides in deep mines has yet to be demonstrated, however.

The uses of bacterial bioreactors to treat heavy-metal-containing industrial wastewaters has been suggested by Gadd (1990), although such techniques could be equally applicable to mine waters (Banks 1992). These reactors may be based on heavy metal-immobilising bacteria (e.g. *Gallionella*) or on sulphate-reducing bacteria.

Conclusion

This paper has reviewed a variety of circumstances in which mine-water drainage can arise and the similar variety in mine-water chemistry that has been observed. Mine waters from apparently similar mine types can be highly acidic or alkaline, depending on the complex interplay of hydraulic, chemical and biological processes occurring. Mine waters may be valuable in terms of high quality baseflow or drinking-water sources; they may be regarded as sources of raw materials; they may be highly ecotoxic and be responsible for devastating watercourses or they may be highly visible, defacing our estuaries and rivers. Unfortunately it is often only the "ugly" mine-water occurrences that wake the attention of the public and politicians. The benevolent mine waters may not be adequately exploited, nor the dangers from the less-visible, but highly ecotoxic, mine waters be adequately appreciated.

A quantitative understanding of the processes governing acid mine drainage remains elusive. These are particularly complex to tackle, not least because the chemistry of mine waters is seldom controlled purely by thermodynamic equilibria between mineral and dissolved phases. A satisfactory quantitative model needs to address the following issues:

1. Three-phase interaction between mineral, gas and water phases, often on a seasonally fluctuating basis (e.g. in the zone of water-table fluctuation).
2. The importance of kinetic factors; the rates of reaction and consumption of carbonate, sulphide and silicate phases (Strömberg and Banwart 1994, Strömberg and others 1994), diffusion-limited steps.
3. The role of bacteria as biocatalysts in sulphide oxidation.

   Nevertheless, a thorough understanding of the mechanisms and controls on the geochemistry of mine-water evolution remains a priority task. There is an acute need to be able to predict the quality of pumped mine water when a new mine is commissioned or of drainage water when a mine is abandoned. Additionally, the chemical nature of a mine water will determine which treatment techniques are likely to be successful.

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