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Long-term changes in the quality of polluted minewater discharges from abandoned underground coal workings in Scotland

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Abstract

Long-term trends of changes in the quality of water discharging from abandoned coal mines have been studied for 32 long-established discharges in the Midland Valley of Scotland. Six discharges have been studied in greater detail, providing insights into discharge quality evolution over more than a century. It has been found that minewater pollution is most severe in the first few decades after a discharge begins (the 'first flush'), and that even the largest systems settle down to a lower level of pollution (particularly in terms of iron concentration) within 40 years. Long-term iron concentrations of less than 30 mg/l are typical, and many are less than 10 mg/l. Low pH values (which might justify the over-used term 'acid mine drainage') do not generally persist, due to the rapid buffering of localized acidic waters by carbonates (both natural carbonates, and those introduced as rock powder for fire precautions during mining). This is corroborated by alkalinity concentrations, which tend to be highest in the early years of a discharge. While the pyrite content of the worked sequence strongly influences initial water quality (in terms of pH, iron and sulphate) during the 'first flush', there appears to be no correlation between long-term iron concentrations of discharges and pyrite content of local strata. Rather, higher levels are found in any sequence where there is scope for fluctuations of the water table in worked ground near to the discharge. A scientific approach to minewater remediation should allow for active treatment of discharges for the first decade or two, followed by long-term passive treatment after asymptotic pollutant concentrations are attained.

Keywords: abandoned mines, acid mine drainage, coal mines, polluted water, water quality

Introduction

An ability to predict the future is one of humanity's greatest urges, and also one of its greatest frustrations. In the context of predicting the likely hydrological consequences of the cessation of regional mine dewatering in large, inter-connected systems of underground workings, the frustrations are often grave. As Sherwood & Younger (1994) have pointed out, prediction of minewater flow patterns in abandoned underground workings is as often hindered by a complete lack of data as by an overwhelming quantity of unintelligible plans. When one turns to the prediction of minewater quality, a complete absence of a reliable archive of hydrochemical analyses is usually the most critical problem. For although mine operators often gather large volumes of data on minewater quality during working, it is generally not a legal requirement for the analytical results to be maintained on public archives. Public mine-records offices are therefore useful sources of physical data (i.e. mine layout) for abandoned mines, but seldom yield any useful subsurface water quality data. Even where analytical results are available for subsurface samples collected during mining, the interpretation of these analyses requires careful consideration of the local stratigraphy and mining history (e.g. Younger *et al.* 1995).

Perhaps most frustrating is the fact that waters sampled during mining are often quite different in quality from those which finally emerge at the Earth's surface after a mine floods (Robins 1990; Younger 1994). This is because the process of flooding brings water into contact with portions of the mine that were formerly dry, as the water 'make' of the mine passed them by in well-worn flow-paths (Younger 1993). The formerly dry areas may contain substantial accumulations of ferric and ferrous hydroxy-sulphate salts (often precipitated as evaporite minerals over many years, in response to forced ventilation) which dissolve readily in the rising water. This dissolution process has been termed a 'geochemical trauma' by Younger (1998). The water which undergoes this geochemical trauma may well be hundreds of times more mineralized than the formerly pumped waters, and in particular may be notably more acidic and ferruginous (Chen *et al.* 1997). Even where waters were acidic and/or ferruginous during working, however, archive analyses of mine effluent samples held by environmental regulators (e.g. the United States' Environmental Protection Agency (US EPA), or the Environment Agency (EA) in England and Wales and the Scottish Environment Protection Agency (SEPA) in Scotland) may be unrevealing, as such waters have often received extensive treatment prior to discharge from the mine premises.

The chemical signature of a discharge when it emerges at the Earth's surface is a result of the dissolution of the products of iron pyrite oxidation and the subsequent chemical reactions that follow. This overall process can be summarized by the equation:

$$4FeS_2 + 15O_2 + 14H_2O \rightarrow 4Fe(OH)_3 + 8SO_4^{2-} + 16H^+.$$

It is clear from this equation that considerable proton acidity is generated under these conditions. Indeed, many discharges are highly acidic upon their first emergence. The well documented discharge from Dalquharran in Ayrshire emerged in 1979 with a pH of 4.1 and iron and aluminium concentrations of 1200 mg/l and 100 mg/l respectively. At the Lower Ynysarwed discharge in South Wales, an initial pH of 3.2 was recorded when it began to flow in 1993.

The impact of such discharges on receiving watercourses is frequently severe. In general, where iron concentrations in the discharge are in excess of 1.0 mg/l, ferric hydroxide (ochre) precipitates as the pH of the metal laden water is increased by the neutralizing effect of the receiving waters (NCB 1982). Precipitation at discharge iron concentrations of below 1.0 mg/l has however, been reported by Ranson & Edwards (1997) in South Wales with the consequence that discharge consent standards are now set by the EA in South Wales such that the iron concentration in the receiving watercourse will be less than 0.5 mg/l.

The ferric hydroxide precipitate can smother the benthos and prohibit photosynthesis along the affected stretch of water. As a result, damage to, or wholesale elimination of, a vital part of the food chain may occur. This is particularly true if initial iron concentrations are sufficiently high (generally in excess of 5 mg/l) (NCB 1982). The direct toxic effects of dissolved iron and zinc may also damage the diversity of the fauna although this has been shown to have a less significant impact than ochre precipitation (Jarvis & Younger 1997).

After a surface discharge commences from a mine, the water quality usually improves over time (Robins 1990; Robins & Younger 1996; Younger 1997*a*). However, there have been very few studies undertaken to evaluate the degree of improvement or its time-scale. Frost (1979) developed a model for this process based on the assumption that mine discharges are dependent on a convective mass-transfer process. The model predicts an exponential decline in concentration of oxidized products in a discharge. Frost (1979) suggests that, provided the concentration of Fe or SO_4 is not controlled by mineral solubility factors, maximum minewater concentrations will be limited by the rates of release of Fe from the initial oxidation sites. (This is somewhat at odds with the classic findings of Singer & Stumm (1970) who suggest that the oxidation of ferrous iron to ferric iron is the rate-limiting step of the overall pollution-generating reaction). In support of his model, Frost (1979) illustrates the exponential decrease in iron concentration at a mine in County Durham, England, in which a partial flooding of voids was arrested by renewed pumping. As the water levels were held steady, the iron concentration declined with a half-life of 350 days. However, it is crucial to note that the model of Frost (1979) holds only for a 'constant volume of flooded workings' i.e. the exponential decline in concentrations modelled and observed are the result of fixed water levels which are artificially maintained by pumping. The evolutionary chemical pattern of a discharge from an abandoned mine, where groundwater levels are not kept at a constant depth but fluctuate seasonally, is unlikely to exhibit such a simple trend; this is because the rise and fall of the water table can flush out further products of pyrite oxidation (Younger 1997*a*). Hence it is not surprising to find that Frost's (1979) optimistic prediction that 'chronic pollution from abandoned mines ... should persist no longer than five years' is flatly contradicted by experiences in County Durham and elsewhere, where some polluting discharges have already been flowing for more than a century.

Glover (1983) took a less theoretical approach to the problem. Drawing upon his extensive experience of water control for the former UK National Coal Board, he suggested that the iron concentration of a discharge from a flooded abandoned mine will halve in each subsequent period of time equal to that taken for the abandoned workings to fill with water after the pumps were withdrawn.

Jones *et al.* (1994) describe how certain minewaters in Pennsylvania, USA, changed from acidic to circumneutral over a period of decades, ascribing the change to the effects of declining ionic strength.

Younger (1997*a*) reviewed the processes likely to control the long-term evolution of the quality of discharges from abandoned mines, concluding that an initial exponential decline (the 'first flush'), the duration of which is determined by the size of the interconnected system of mined voids in relation to the recharge rate, would be followed by an asymptotic level of pollution, controlled by the degree of ongoing pyrite oxidation (or, in some cases, siderite/ankerite dissolution) in the zone of water table fluctuation (Strömberg & Banwart 1994).

Consideration of the processes that can occur within the abandoned mine environment highlights a number of time-*independent* factors and a number of time-*dependent* factors which may affect discharge quality, and hence the position of a given minewater on its hydrochemical evolutionary path.

(1) The *time-independent* factors are:

- The *lithological setting* of the worked coal—coals associated with strata of a marine/brackish water origin are likely to contain more abundant oxidizable framboidal pyrite than those associated with non-marine strata and thus produce more polluting water (Caruccio & Ferm 1974).

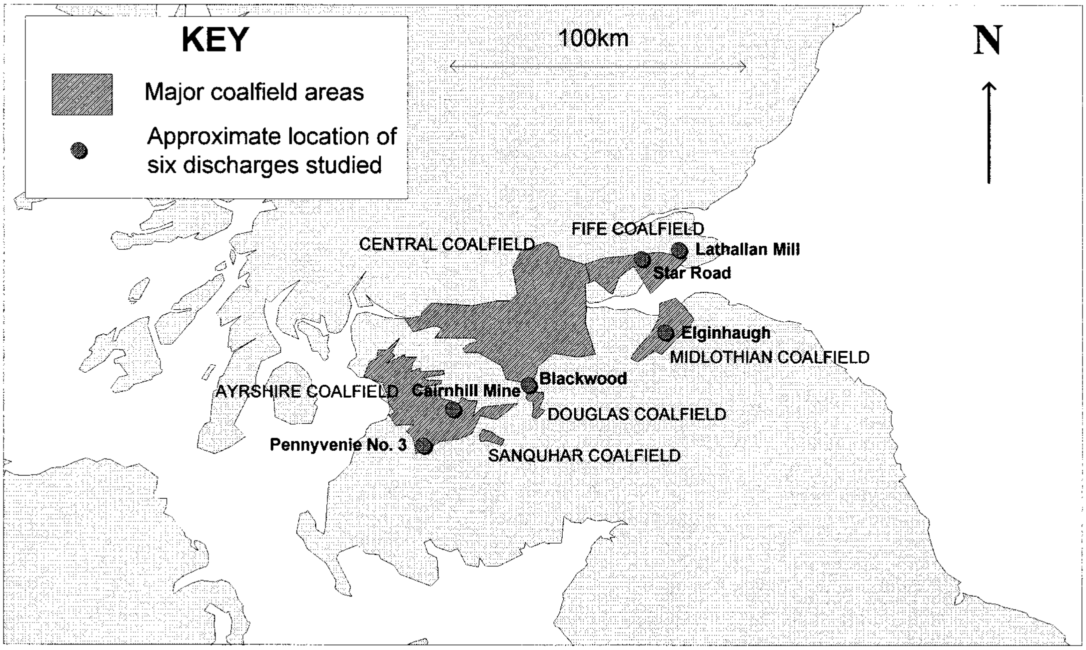

Fig. 1. Locations of six discharges studied in relation to the Scottish coalfields.

- The *extent of coal workings*—the greater the extent of coal workings the greater the surface area of oxidizable pyrite and thus the emergence of more polluted waters from large interconnected mining areas.

(2) The *time-dependent* factors are:

- The transition from '*vestigial acidity*' (Younger 1997*a*) (which is acidity generated from the flushing process during water table rebound) to '*juvenile acidity*' (which is acidity generated from the seasonal fluctuation of the water table when it has recovered an equilibrium position after being depressed). Juvenile acidity should, in theory, be far less concentrated than vestigial acidity in that seasonal pyrite oxidation and subsequent flushing will only occur within the zone of water table fluctuation.
- *carbonate dissolution* after acid generation—this arises from associated limestone horizons or artificially-introduced limestone in the workings, and may effectively buffer the pH and result in significant alkalinity.
- The *iron hydroxide 'armouring'* of pyrite surfaces at pHs above 4. This occurs by the deposition of insoluble iron hydroxide on pyrite surfaces, which provides an oxygen barrier and hinders further pyrite oxidation. This cumulative process will, in effect, reduce the iron and sulphate concentration in solution and reduce the acidity.
- The *bacterial reduction of sulphate*—this reduces the sulphate in solution and generates bicarbonate alkalinity. It is likely to be time dependent as the bacteria will only become established in anoxic conditions which probably do not develop until water table rebound has occurred.
- The *change in water flow rate* through the mine—this may result in the concentration or dilution of the chemical constituents in the discharge.

It is the purpose of this paper to examine the magnitude and timing of these factors by studying the long-term evolution of water quality in many of the abandoned coal mines of the Midland Valley of Scotland.

Study Methods

All available water analyses for 32 (out of a total of more than 80) existing abandoned coal mine discharges within the Forth and Clyde catchments (including those for the well documented discharges at Dalquharran and Fordell Castle) were collated from records now held by SEPA. The age of each individual analysis was determined in terms of the time since the abandonment (or, where recorded, the end of local dewatering) of the associated coal mine.

Various parameters, for all included discharges, were then plotted as a function of 'discharge age' to ascertain the existence of any broad evolutionary trends characteristic of coal mine discharges in general.

Six discharges were then selected from the complete data set with ages ranging from 17 years to 116 years old

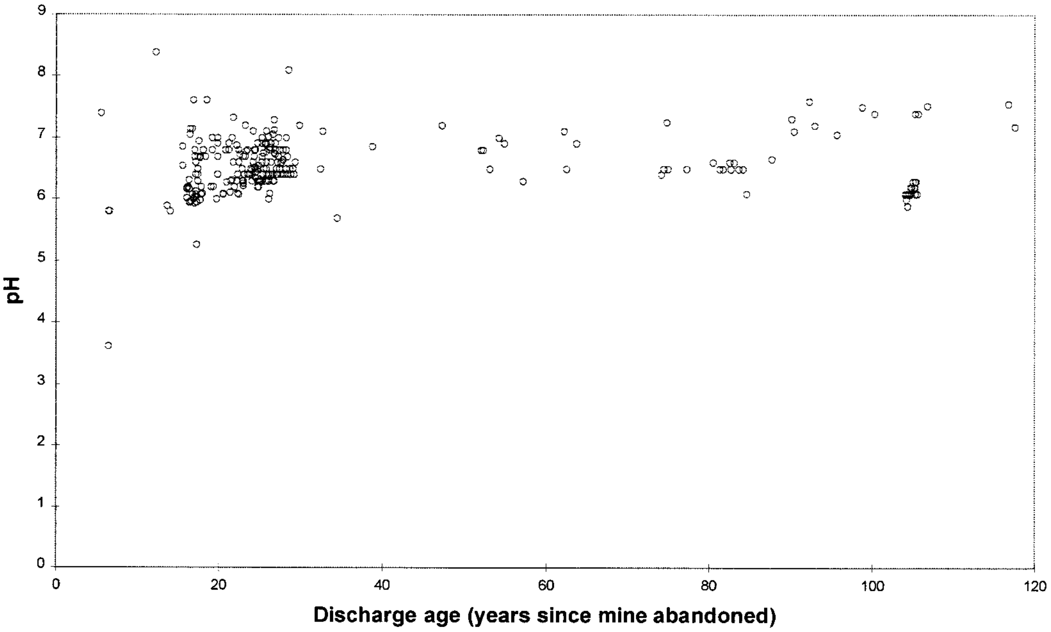

Fig. 2. Variation in pH with age of discharge (for all discharges).

(see Fig. 1 for discharge locations). This was the number of discharges that could be adequately investigated within the study period. Selection was governed by the following factors:

- A relatively even spread of discharge ages was required over a broad age range.
- The mineworkings were ideally required to be discrete, isolated systems in order that complications of hydraulic connections to other mines were avoided and that the impact of void space could be assessed.

These discharges were analysed in more detail to discover how site-specific complicating factors may manifest themselves within any of the broad evolutionary trends observed. Each discharge was sampled in the field to obtain a more comprehensive chemical analysis than was previously available and the geological setting of each mined environment was investigated using archival records held by the British Geological Survey (BGS) at Murchison House, Edinburgh, and by the Coal Authority and other organizations.

The study area

The discharges under scrutiny are associated with the mined strata within the Midland Valley of Scotland. Geologically, the Midland Valley is a graben, largely filled with Devonian and Carboniferous rocks, and is bounded by the Highland Boundary Fault to the north and the Southern Upland Fault to the south (Craig 1991).

The Devonian sequence is composed of conglomerates, sandstones, siltstones and mudstones with sandstone the dominant lithology. The Carboniferous system is represented by rhythmic sequences of coals, limestones, mudstones, siltstones, sandstones, seatearths and seatclays which vary significantly in thickness laterally. It is in the Carboniferous lithostratigraphic divisions known as the Lower Limestone Group, the Limestone Coal Group, the Upper Limestone Group and the Coal Measures that the majority of coal mining activity in Scotland has been concentrated (Francis 1991). The geographical location of these coalfields is illustrated in Fig. 1.

The first references to coal mining in Scotland date from the end of the 12th century in monastic charters. From those primitive beginnings, when coal seams were worked from outcrop, a sophisticated industry developed, which has resulted in just short of 20 000 documented shaft entrances to deep mines in Scotland. Today, however, only one of these mines (Longannet) remains in operation. It is the closure of large numbers of mines (over 200 deep mines since nationalization in 1947) that has led to the appearance of more than 80 polluted minewater discharges in the area (Henton 1981; Robins 1990; Robins & Younger 1996; Younger *et al.* 1995; Chen *et al.* 1997). The impacts of these discharges are serious. For instance, in the Forth catchment, discharges from abandoned mines are now the single greatest cause of freshwater pollution (FRPB 1995).

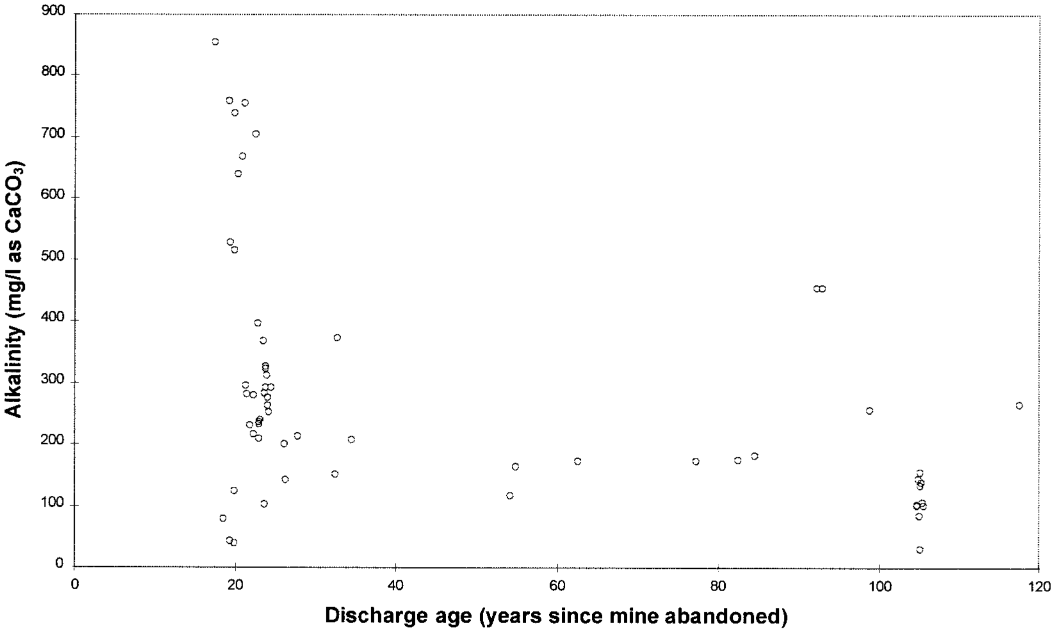

Fig. 3. Variation in alkalinity (mg/l as $CaCO_3$) with age of discharge (for all discharges).

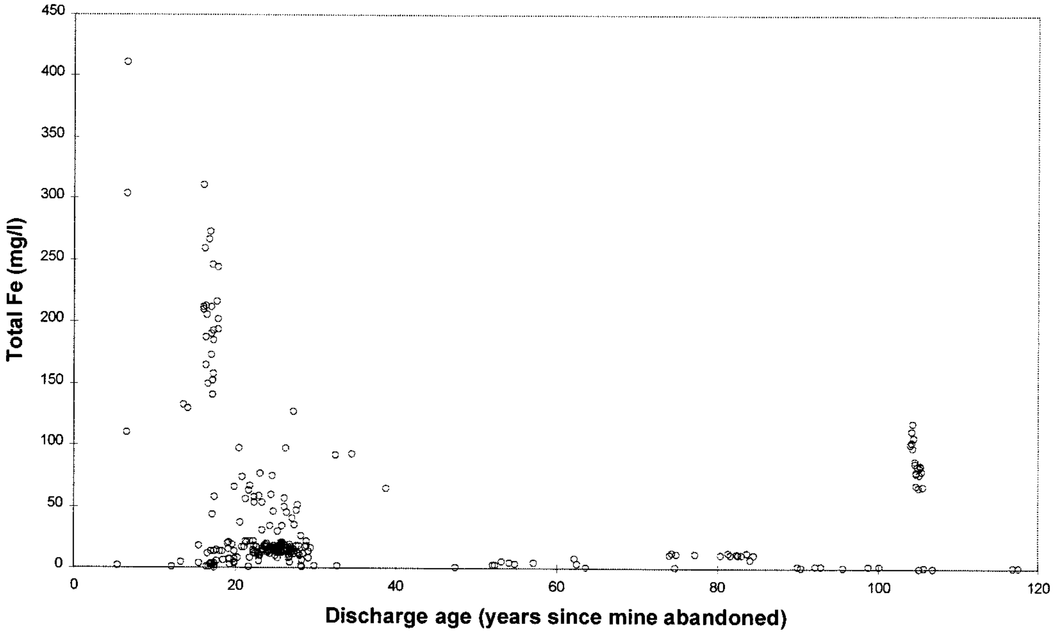

Fig. 4. Variation in total Fe (mg/l) with age of discharge (for all discharges).

Results

Figures 2–5 show all the available data for the key analyses plotted, for 265 samples from 32 discharges. Results of the six detailed site studies are given in Tables 1 and 2, and in Figs 6–9.

Discussion and Interpretation

The broad trends revealed by the available data for all 32 sites are as follows. Figure 2 reveals that none of the discharges within the data set has a pH that falls even close to 'acid mine drainage' values (which can be as low

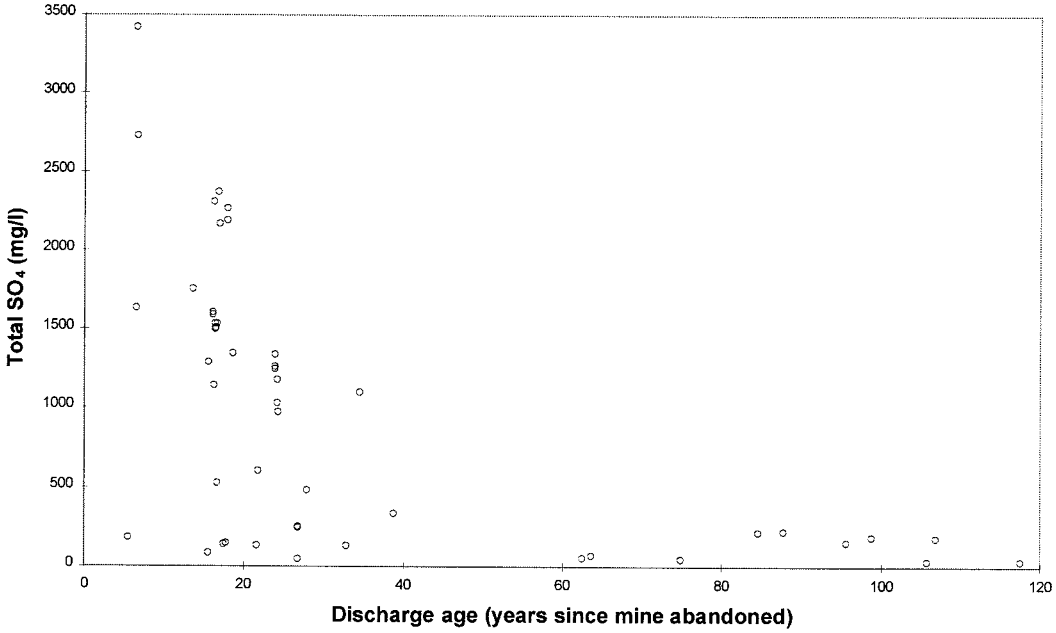

Fig. 5. Variation in sulphate (mg/l SO_4) with age of discharge.

as 2). Rather, all of the discharges appear to reach a circum-neutral state within approximately 30 years of emergence. Perhaps also surprisingly, alkalinity is highly variable, but generally relatively high for approximately 25 years from the emergence of flow (Fig. 3), declining thereafter to fluctuate around approximately 200 mg/l. There are two obvious explanations for this phenomenon. Firstly, it was common practice in Scottish deep mines (as in northern England) to spread limestone slurry ('rock powder') around the walls of the mine to provide a 'flame blanket' to prevent ignition of coal dust. Upon flooding, this limestone cake is readily available for dissolution by initially acidic waters. Secondly, if waters were initially highly acidic, reaction with carbonates present naturally in the rock will, provided the supply of carbonates is not limited, result in a correspondingly high level of dissolved HCO_3 after neutralization. Unfortunately, SEPA records for calcium in the minewaters are too sparse to allow corroboration of this process by cross-plotting of calcium and bicarbonate.

Iron concentrations appear to be wide-ranging in the first 40 years of outflow (0–400 mg/l) but after that are below 30 mg/l (with the exception of one cluster of data at 105 years which corresponds to discharge analyses from a single mine) as shown in Fig. 4. Similarly, sulphate concentrations are restricted to a much lower range after approximately 40 years of discharge flow (Fig. 5).

As the predominantly circum-neutral pHs illustrated in Fig. 2 demonstrate, this declining trend is unlikely to be simply related to pH. The relationship between pH and iron concentration is not straight-forward due to the multiplicity of 'sink' reactions which operate when a water is very aggressive. Circum-neutral waters can have Fe concentrations as high as 250 mg/l while net-acidic waters can have Fe concentrations up to 2500 mg/l but, because of these sink reactions, concentrations are often as low as 20 mg/l.

The trend is more likely to be governed by the mechanism of water table rebound and its interaction with oxidized pyrite. The widely ranging concentrations observed in the first 40 years are likely to be a result of the different durations of water table recovery. Iron and sulphate concentrations and acidity will fall at a rate which is a function of the rate of dilution. The rate of dilution will be greatest where the volume of mineworkings is small, the hydraulic conductivity of the mined system is high and the recharge rate to it is high (Younger 1997*a*).

Detailed site studies for the six discharges listed in Table 1 corroborate the trends seen in the 32 discharges, but also provide an indication of the extent to which the various time-dependent and time-independent factors (identified above) affect the quality of each discharge. A summary is presented in Table 2.

Figure 6 suggests that after ten years of flow, pH may become circum-neutral irrespective of:

(i) the origins of the associated source strata and hence the abundance of pyrite, and
(ii) the presence or absence of limestone beds which may buffer the groundwater.

Table 1. *Analysis of minewaters at six discharges studied in detail*

	Lathallan Mill	Star Road	Elginhaugh	Blackwood	Cairnhill	Pennyvenie No. 3
NGR	NO 465 063	NO 296 025	NT 317 670	NS 803 432	NS 627 234	NS 487 066
Sample date	9/7/95	9/7/95	9/7/95	10/7/95	10/7/95	10/7/95
Flow rate (l/s)	16	3	approx. 55	2.6	0.09	6
Temperature (°C)	11.3	11.4	13.8	9	17.9	14.3
pH	6.1	6.5	5.7	7.2	7.6	6.9
Eh (mV)	17	−57	6	−40	−76	−7
Conductivity (mS)	0.82	0.47	0.78	0.42	1.96	1.75
Alkalinity (mg/l $CaCO_3$)	182	173	207.5	265.5	79.5	854.5
Acidity* (mg/l $CaCO_3$)	20.8	9.8	191.6	3.4	27.4	2.3
Mn (mg/l)	0.70	0.54	11.52	0.29	4.02	0.53
Al (mg/l)	0.04	0.20	0.59	0.16	1.10	0.15
Zn (mg/l)	0.07	0.02	0.41	0.01	0.05	0.01
Total S (mg/l)	66.29	15.21	394.00	12.52	408.80	46.36
Ca (mg/l)	87.03	61.00	256.40	75.48	384.95	57.98
Mg (mg/l)	41.32	21.391	88.10	26.81	88.00	42.05
Na (mg/l)	16.17	13.08	15.94	16.93	27.99	302.55
Total Fe (mg/l)	10.81	4.02	92.78	0.71	6.74	0.26
Fe^{2+} (mg/l)	10.81	3.40	91.00	0.00	4.55	0.26
Fe^{3+} (mg/l)	0.00	0.62	1.78	0.71	2.19	0.00
Si (mg/l)	7.68	5.64	7.13	5.34	10.49	4.86
Ni (mg/l)	0.01	0.00	0.26	0.01	0.04	0.00
K (mg/l)	5.91	12.24	22.83	12.24	71.97	24.37
Fl (mg/l)	0.28	0.20	1.58	0.18	0.78	0.75
Cl (mg/l)	34.89	33.76	21.46	29.70	23.05	21.40
SO_4 (mg/l)	214.40	52.83	1101.39	36.77	1346.13	141.64
NO_4 (mg/l)	0.13	0.00	0.00	0.00	15.62	0.87
Br (mg/l)	0.00	0.00	0.00	0.00	0.78	0.12

*Acidity values are calculated from the equation given by Hedin *et al.* (1994).

Table 2. *Summary of investigation of six discharges studied in detail*

Discharge	Age (years)	Time-independent factors		Time-dependent factors				
		Extent of workings	Associated coal lithology	Vestigial or juvenile phase	Source of carbonate	FeOOH armouring (supersat'd/ undersat'd)	Sulphate reduction	Flow rate
Lathallan Mill	approx. 100	Small scale	Marine	Juvenile	Present	Unlikely (undersat'd)	Likely	—
Star Road	approx. 106	Small scale	Marine	Juvenile	Absent	Likely (supersat'd)	Unlikely	—
Elginhaugh	35	Very extensive	Freshwater	Uncertain	Present	Likely (supersat'd)	Likely	Fairly constant
Blackwood	117	Small scale	Marine	Juvenile	Absent	Likely (supersat'd)	Likely	—
Cairnhill	20	Medium scale	Marine	Uncertain	Absent	Likely (supersat'd)	Likely	Much reduced
Pennyvenie No. 3	18	Extensive	Freshwater	Uncertain	Absent	Likely (supersat'd)	Likely	—

It would therefore appear that acid generation is inhibited after approximately ten years of flow for the six study systems. This is likely to be the result of a combination of the change from vestigial to juvenile acidity and iron hydroxide armouring of pyrite surfaces within the zone of seasonal water table fluctuation.

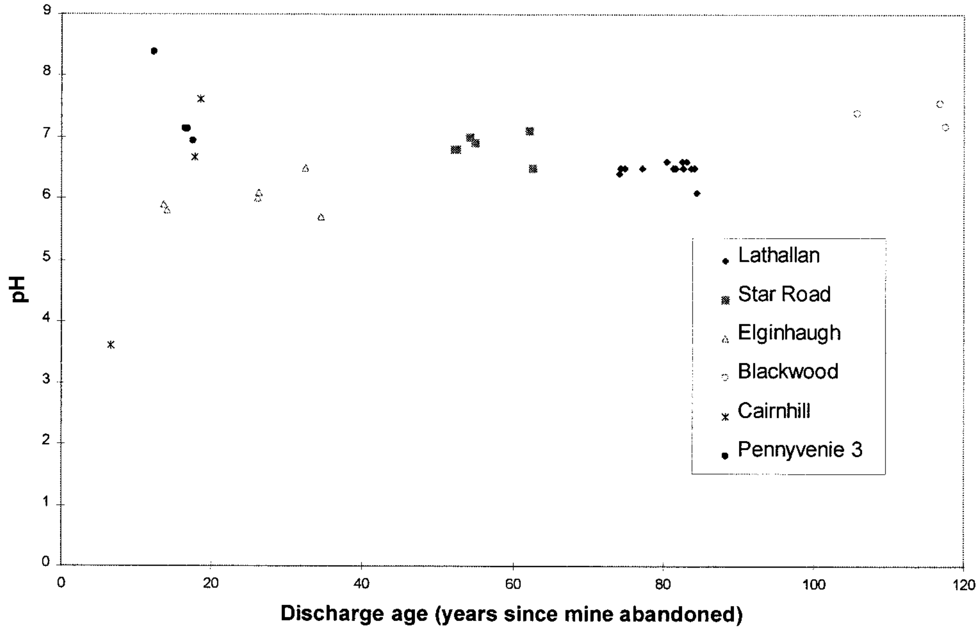

Fig. 6. Variation in pH with age of discharge for six detailed study sites.

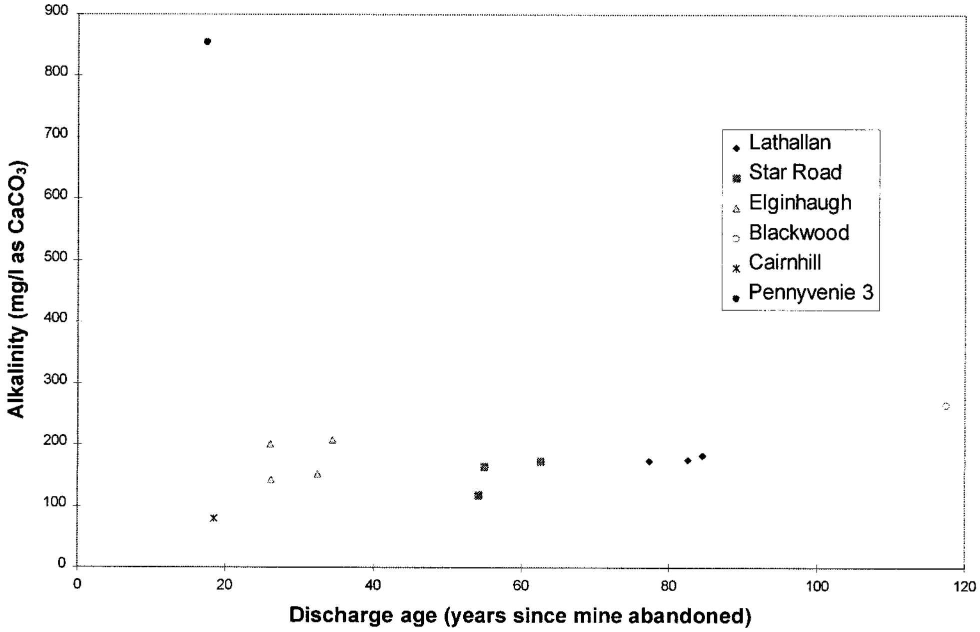

Fig. 7. Variation in alkalinity (mg/l as $CaCO_3$) with age of discharge for six detailed study sites.

Figure 7 reveals no clear trend in alkalinity. The high alkalinity exhibited by the Pennyvenie 3 discharge cannot, it seems, be attributed to a lithological carbonate source as Table 2 shows. However, the very high molar ratio of HCO_3^- to $(Ca^{2+}+Mg^{2+})$ suggests that significant bicarbonate generation by bacterial sulphate reduction may be occurring since a molar ratio of approximately 1.00 would be expected if both species were derived from calcite or dolomite dissolution. Nevertheless, Fig. 3 would suggest that a cut off point exists at between 20 and 30 years beyond which particularly high alkalinities are not experienced. This may be

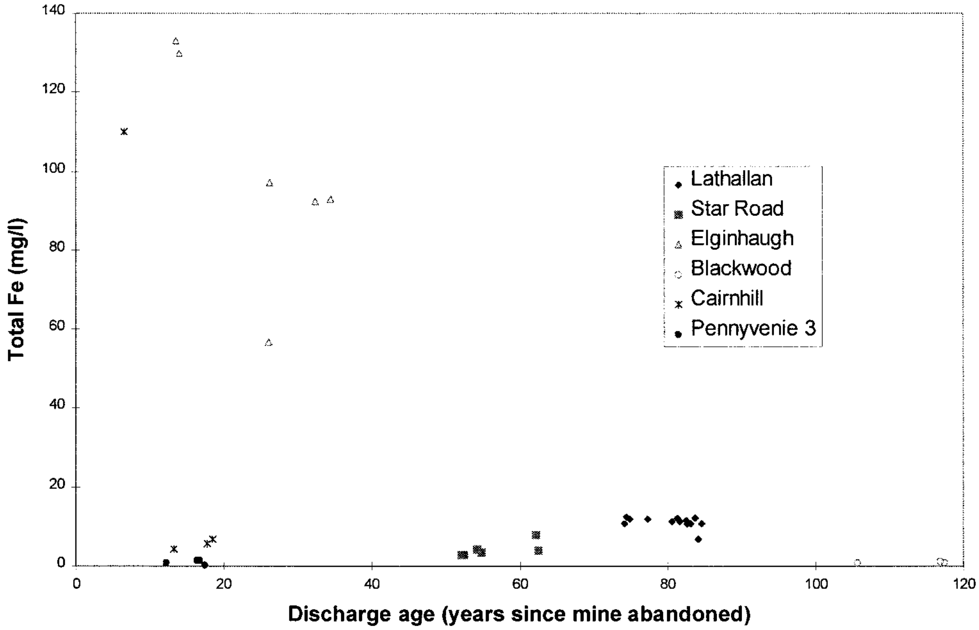

Fig. 8. Variation in total Fe (mg/l) with age of discharge for six detailed study sites.

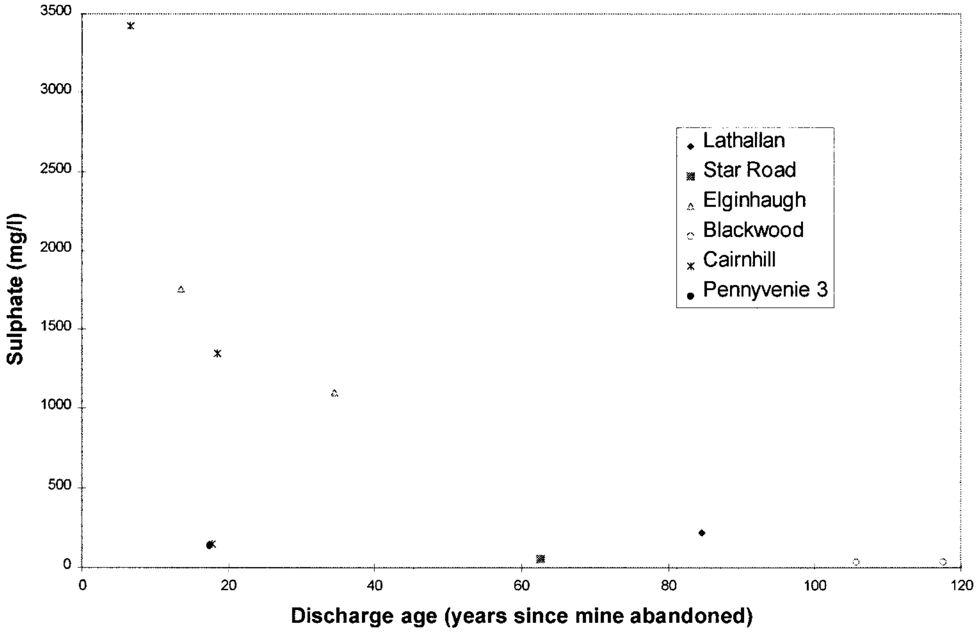

Fig. 9. Variation in sulphate (mg/l SO_4) with age of discharge for six detailed study sites.

due to the exhaustion of limestone powder within the workings. Further investigation of typical rock powder application rates might shed light on this.

There appears to be no direct relationship between extensive workings and high iron and sulphate concentrations, nor between discharge age and high iron and sulphate concentrations for discharges which have been flowing for less than 40 years. This is supported by the plots for Elginhaugh and Pennyvenie 3 (Figs 8 & 9), both of which are relatively young discharges emerging from extensive workings. What does appear evident, however, is that discharges that have been flowing for more than 40 years or so exhibit much reduced iron and sulphate concentrations.

Implications for minewater pollution remediation

The findings of this study demonstrate quite clearly that an over-emphasis in the long-term on the acidic nature of deep mine discharges is unwarranted. In the vast majority of cases, the key problem will be iron, and pH control will be required for a relatively short period after minewaters start to flow, possibly up to ten years. However, this is not the case in relation to discharges from shallow, drift mines and spoil heaps where passive ventilation facilitates more vigorous pyrite oxidation in the shallow subsurface and can sustain low pH values for decades (Younger *et al.* 1997). As the loadings of all pollutants decline markedly over time, it would make sense to plan for active treatment of new discharges from abandoned mines using proven technologies such as lime-dosing and sedimentation, only for the first decade or so (Younger 1997*a*). Even the largest mine systems will reach asymptotic quality after four decades. Thereafter, low-cost, low-maintenance solutions using constructed wetlands (Hedin *et al.* 1994; Younger 1995*a*,*b*, 1997*b*; Younger *et al.* 1997) may be designed with minimal land-take and minimal process sophistication (since wetlands designed to treat ferruginous, circum-neutral waters have to be far less elaborate than those treating acidic waters).

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