The operational performance of Scottish minewater-based ground source heat pump systems

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Abstract: Two open-loop, minewater-based, ground source heating schemes have been operating since 1999–2000 in Scotland (UK), at Shettleston (Glasgow) and near Cowdenbeath (Fife). In both cases, ferruginous water is pumped from flooded coal mine workings via a borehole, circulated through the evaporator of the heat pump and reinjected via a shallower borehole to Carboniferous strata. The heat is delivered to a water-based thermal store providing central heating for apartment complexes and contributing to domestic hot water. It is demonstrated, via hydrochemical analysis and speciation modelling of the Fife minewater, that the success of the schemes is due to lack of contact between minewater and the atmosphere (thus limiting degassing of CO_2 and absorption of O_2). Indeed, recent difficulties with one of the schemes are ascribed to vandalism of the recharge main allowing access to oxygen, causing precipitation of ferric oxyhydroxide and clogging of the recharge borehole.

This paper is one of a thematic set entitled 'Hydrogeology and Heat Engineering'. The use of groundwater, abstracted via open-loop systems, is increasingly being considered as a means to provide heating and cooling for buildings, particularly in London. To avoid unnecessary repetition between papers in the set, the background to the science of thermogeology and the exploitation of ground source heat has been described by Banks (2009) and the UK regulatory environment by Fry (2009).

Heat pump technology allows us to extract low-temperature ambient heat from the environment, to upgrade it to high-temperature heat and to use it for space heating. Heat pumps that extract heat energy from the geological environment (rocks, soil, sediments or groundwater) are typically referred to as ground source heat pumps (GSHP). Such GSHPs do not require pristine, uncontaminated ground conditions: heat can be extracted from contaminated soils or contaminated groundwater, providing an excellent way of 'rebranding' a polluted site as a source of 'renewable energy'.

One of the most attractive potential applications for GSHP technology in Britain is the extraction of heat from minewater from abandoned (or even active) mines. The UK Coal Authority is currently responsible for managing in excess of 3000 l s^{-1} water from over 50 abandoned coal mine sites across England, Wales and Scotland. Assuming a coefficient of performance of 3.5 and a temperature drop at the evaporator of 5°C, heat pump systems could deliver c. 88 MW of space heating power on the basis of this water flux:

$$\frac{3000 ls^{-1} \times 4180 Jl^{-1}K^{-1} \times 5K}{\left(1 - 1/3.5\right)} \approx 88 MW \quad (1)$$

Similarly, in the vicinity of Katowice, Poland, the state mine dewatering organization CZOK pumps an average of 2655 l s^{-1} water from abandoned coal mines of the Upper Silesian Coal Basin, representing a potential space heating effect of 78 MW (Janson *et al.* 2009).

Such minewater discharges are usually regarded as potential sources of pollution because of the typically elevated concentrations of dissolved iron, manganese (and sometimes aluminium) and sulphate resulting from the pyrite oxidation reaction:

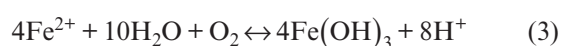$$2FeS_2 + 2H_2O + 7O_2 = 2Fe^{2+} + 4SO_4^{2-} + 4H^+_{(aq)}. \quad (2)$$
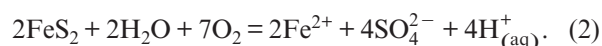
Indeed, the UK Coal Authority has a rolling programme of constructing minewater treatment schemes at the most potentially polluting coal mine discharges. Although such minewater treatment schemes are often based on wetlands and are designed to be 'passive' as far as possible, all of them will have continuing maintenance requirements, some of them incur continuous pumping costs and a few of them require active chemical dosing (Banks & Banks 2001). They thus represent a continuing environmental liability for the Coal Authority and incur a continuing operational cost. However, many former colliery sites are now being redeveloped as commercial or residential sites (and some as museums), all with heating and cooling requirements. The possibility of using minewater as a source of heat and 'coolth' for such new developments offers the Coal Authority an
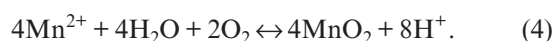
opportunity, not only to parade its environmental credentials by converting 'orange minewater' to 'green energy', but also to recoup some of its operating expenditure by selling heating and cooling services (Table 1). In Britain, the underground mining industry is almost defunct, but in operational mines, heat could feasibly be extracted from mine wastes or minewaters to heat mine buildings, pithead bath-houses or even to regulate the temperature and humidity of ventilation air.

The idea of extracting heat from minewaters is not new. Banks *et al.* (2004) have demonstrated that the concept is feasible and that it has been applied in practice in the USA, Canada and Norway. Further feasibility studies and compilations of case studies have been published by Watzlaf & Ackman (2006) and Fraga Pumar (2007). In the Netherlands, a pilot project is under way to provide district heating and cooling from coal minewater in the municipality of Heerlen (http://www.minewaterproject.info/). In Poland, feasibility assessments have reached a relatively advanced stage (Solik-Heliasz & Małolepszy 2001; Małolepszy *et al.* 2005), in part because of the relatively high temperatures of some of the waters pumped from deep mines in the Upper Silesian Coal Basin (>20°C from Gliwice, Szombierki, Siemianowice, Dębieńsko and Powstańców Śląskich mines; Janson *et al.* 2009).

In fact, the UK and Poland have ideal economic and geological conditions to develop minewater as a renewable energy source. We must therefore ask: why haven't the mining authorities or private site owners actually built such a mine-sourced heating scheme yet? One reason is possibly related to the observation that coal minewater is prone to precipitation, on the beds or rivers and in wetlands, of iron and manganese oxyhydroxides by the reactions

$$4Fe^{2+} + 10H_2O + O_2 \leftrightarrow 4Fe(OH)_3 + 8H^+ \qquad (3)$$

and

$$4Mn^{2+} + 4H_2O + 2O_2 \leftrightarrow 4MnO_2 + 8H^+. \qquad (4)$$

It is feared that this same hydroxide precipitation reaction may also take place within the heat exchangers of a heat pump system, causing rapid clogging and the need for an onerous programme of maintenance.

Such 'clogging' and a concomitant occasional maintenance requirement has been documented in the case of some open-loop GSHP schemes, based on 'normal' groundwater, such as that at Gardermoen International Airport in Norway (Midttømme *et al.* 2008). It may, therefore, come as a surprise that two operational coal minewater heat pump schemes are functioning in Scotland at present and have been doing so (relatively successfully) for 8–9 years.

The development of mine-sourced heating in Scotland

The idea of using minewater in abandoned, flooded collieries as a source of heating was considered in the early 1990s in Scotland. Fortunately, the idea was not left on the drawing board, but was put into practice in a pilot project at Mossend ([NS 74 60], Fig. 1), near Ravenscraig Steel Works, in Lanarkshire in around 1992 by an enthusiastic group of professionals including the engineers S. Johnston (of EnConsult, Dunfermline) and W. Goldie, and the geologist T. Burke. In this pilot scheme, a borehole was drilled to access flooded mine workings and the water was pumped through a heat pump at the surface, with the heat produced being used for space heating in a portable cabin (S. Johnston, pers. comm.). The scheme is no longer in use.

Full-scale minewater-sourced heat pump schemes

The Mossend trial provided confidence that a full-scale minewater-based heat pump scheme would be viable. The group of specialists collaborated closely with John Gilbert Architects of Glasgow to commission, in 1999–2000, two new minewater heat pump schemes at housing association apartment complexes in Shettleston, Glasgow and Lumphinnans, Fife (Fig. 1). The two schemes share several features, which are described in this section. The subsequent sections detail the peculiarities of each scheme and the operational experiences.

The schemes use abstraction boreholes drilled to depths of $c.$ 100 m (Shettleston) and 172 m (Lumphinnans), respectively, and fitted with plain casing and $c.$ 150 mm plastic well screens (S. Johnston, pers. comm.) in the productive horizon. Both boreholes were constructed into flooded coal mine workings from which water is abstracted by an electric submersible pump. The minewater then passes through a steel gauze filter to remove particulate matter before entering the evaporators of the heat pump array. The cooled minewater exits the evaporator(s) of the heat pump(s) and is then reinjected to the ground via a shallower borehole (reported to be typically 50–60 m deep and without well screen; S. Johnston, pers. comm.). In both cases, reinjection is designed to take place through a simple open reinjection main, below the water level in the borehole. In other words, the pump–evaporator–reinjection system is a 'sealed' pipe, and the minewater is not permitted to come into contact with the atmosphere.

At both sites, the heat pump array comprises two ClimateMaster WE120 water-to-water units, using R-22 ($CHClF_2$) refrigerant, with a nominal combined output of up to 65 kW at each location, running on a negotiated cheap 'Eco' electricity tariff. The heat pumps produce hot water at a temperature of around 55°C, which is

Table 1. *Temperatures of minewater discharges from several English abandoned lead and coal/gannister mines*

Location	Grid reference	Discharge	Date	T (°C)	Type
Allen Hill Spaw, Matlock	SK 296 604	0.15 l s⁻¹	20/1/95	10.2	Lead mine adit
Bullhouse	SE 2140 0290	25–30 l s⁻¹	12/7/06	17.9*	Intercepted coal mine adit
Caphouse (Hope Shaft)	SE 2497 1626	76 l s⁻¹ for 16 h day⁻¹	14/6/06	14.2	Pumped discharge from coal mine shaft; 150 m depth to water level, 205 m full depth (water level constant since 2006)
Clough Foot	SD 9051 2378	5–20 l s⁻¹	22/6/06	9.3	Coal mine adit
Deerplay	SD 8644 2754	20 l s⁻¹	22/6/06	8.4	Seepage from near coal mine adit
Duke's Level, Buxton	SK 0370 7231	15–30 l s⁻¹	22/3 to 2/8/95	7.2–7.5	Coal mine canal/drainage adit
Fender, Chesterfield	SK 366 751	20–30 l s⁻¹	31/5/95	9.4	Possibly shaft to Dunston Colliery
			13/6/06	10.0†	
Gawber Prospect	SE 329 086	4.5 l s⁻¹	5/2/95	10.2	Overflowing coal exploration borehole
Goyt Moss, Buxton	SK 018 721	1–2.5 l s⁻¹	22/3 to 2/8/95	6.4–6.7	Coal mine adit
Greystones, Sheffield	SK 3143 8506	0.5 l s⁻¹	31/5/95	9.3	Overflow from abandoned colliery
Horden, Peterlee	NZ 4423 4186	120 l s⁻¹	14/5/08	17.2	Pumped discharge from coal mine shaft; 78 m depth to water level, 406 m to base of shaft
Hunter's Bar, Sheffield	SK 3324 8574	3.3 l s⁻¹	31/5/95	9.8	Overflow from abandoned colliery
Jackson's Bridge	SE 1639 0735	c. 15 to >50 l s⁻¹; highly variable	12/7/06	11.3	Adit from Hazlehead Colliery
Limb Brook, Sheffield	SK 3179 8178	1–2 l s⁻¹	1/2/95	9.0	Shallow gannister and/or coal mine shaft
			13/6/06	9.8	
Odin Sough, Castleton	SK 150 831	4 l s⁻¹	3/1/95	8.7	Lead mine drainage adit
Old Meadows, Bacup	SD 869 242	30–80 l s⁻¹; variable	22/6/06	9.7	Colliery adit
River Hipper, Chesterfield	SK 3842 7067	0.5–0.75 l s⁻¹	22/3/95	10.3	Probably overflow from Boythorpe Colliery drift
			1/5/95	10.7	
Sheephouse Wood	SE 2302 0012	<1 to >16 l s⁻¹, recently c. 6 l s⁻¹	16/1/95	9.2	Adit from Handbank/Hollin Wood collieries
			13/7/06	11.2	
Shepley Dyke	SE 2129 1031	2–3 l s⁻¹	12/7/06	10.3	Colliery shaft
Silkstone	SE 2907 0562	4–5 l s⁻¹	14/6/06	10.2	Colliery adit?
Summerley	SK 3650 7804	10–30 l s⁻¹	13/7/06	10.2	Free-draining discharge from Summerley Colliery
Unstone	SK 3709 7751	12–18 l s⁻¹	13/7/06	11.6	Free-draining discharge from Spring Bank Colliery
Wharncliffe Chase	SK 304 954	c. 0.5–0.75 l s⁻¹	14/6/06	11.7	Adit from gannister/coal mine
Woolley	SE 3064 1144	c. 90 l s⁻¹	22/6/06	15.5	Pumped discharge from coal mine shaft; 105 m depth to water level, 387 m full depth (water level constant since 2006)

It should be noted that the warmest waters (Woolley, Horden and Caphouse) are from deep pumped shafts. Data compiled from Banks *et al.* (1997) and McAllan *et al.* (2009).
* Sampled in treatment works and probably not representative of natural *in situ* conditions.
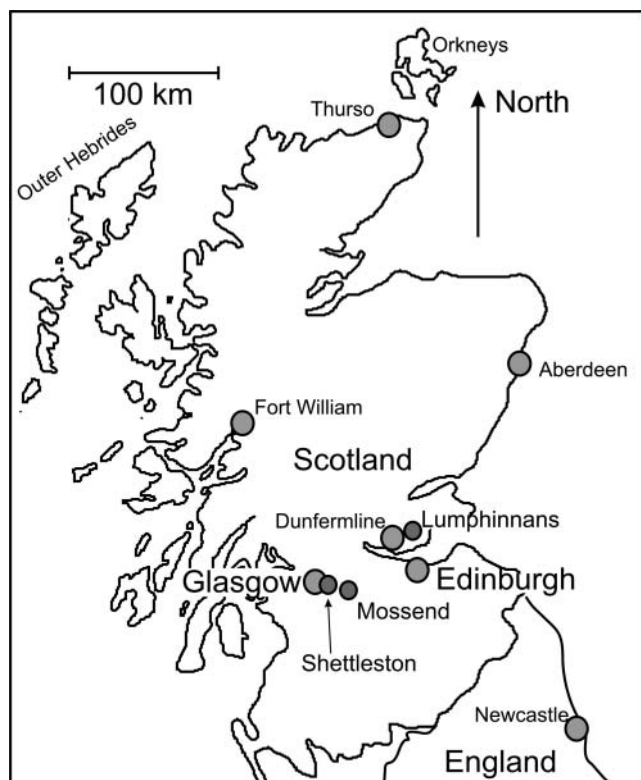† Sampled at inlet to treatment works.

Fig. 1. Outline map of Scotland, showing the localities named in the text.

stored in large insulated thermal storage tanks within the building. From here, water is distributed to provide space heating via radiators and to pre-heat domestic hot water (supplementary electric immersion heaters are used to further raise the domestic hot water temperature). At both Shettleston and Lumphinnans, a portion of the thermally 'spent' minewater from the heat pumps was originally diverted for use as 'grey water' for toilet flushing in the apartments. This was rapidly discontinued at both sites, however, because of oxidation and precipitation of iron from the minewater causing fouling and staining of the toilet systems (SUST 2006).

No routine monitoring of minewater yield or quality takes place, but one would expect that a yield of around 2–3 l s^{-1} would be adequate to service a peak heat load of 65 kW.

Glenalmond Street, Shettleston

The Shettleston Housing Association constructed a new 1600 m^2 complex of 16 apartments in the suburb of Shettleston in eastern Glasgow in 1999 ([NS 644 637], Fig. 2) and the capital cost of the heat pump scheme was covered by a Housing Association Grant.

The Shettleston area is underlain by Middle Coal Measures strata of the Lanarkshire Coalfield, which have been worked locally for coal and fireclay for at least 200 years. Coal has been taken from at least six seams, of which the Upper, the Ell and the Main are the

stratigraphically highest. The 100 m deep abstraction borehole is believed to penetrate abandoned workings in the Ell Seam of Westmuir Pit from the period 1845–1862 (Fig. 3). The pump runs 24 h per day and delivers minewater at an estimated temperature of 12°C (John Gilbert 2000; SUST 2006). The shallower reinjection borehole is located around 20 m from the abstraction borehole. Unfortunately, no documentation is available to confirm whether the reinjection horizon is another mined seam or a sandstone horizon.

The scheme was commissioned in 1999 and there has never been any significant failure of the scheme as a concept. The scheme delivers space heating via low-temperature radiators and is generally favoured by residents in the complex (SUST 2006). An area of 36 m^2 of solar thermal roof panels supplements the temperature of the thermal store, and a back-up electrical heater has subsequently been added to the thermal store in case of problems with the heat pumps (Pither & Doyle 2005). Each apartment pays £90–100 per year for heating and hot water. This figure is designed to cover the electricity consumption of the pump and heat pumps. The housing association acknowledges that it subsidizes the maintenance bill.

As regards maintenance, the in-line filter is replaced every 3 months. The material found in the filter appears to be a mixture of particulates and ochre, the quantities being greatest after heavy rainfall: significant accumulations of ferric oxyhydroxides have not, however, been observed. A sample of the material from the filter was provided for X-ray diffraction (XRD) analysis at Newcastle University (Fraga Pumar 2007), which revealed it to be composed dominantly of quartz, with some calcite, and only minor goethite (although any amorphous iron oxyhydroxide would not have been recognized by XRD). This suggests that the majority of material trapped in the filter comprises clastic particles from the Coal Measures sandstones. The only other maintenance item on the scheme to date has been the replacement of the electric submersible pump after 8 years of service. The heat pump evaporators have reportedly never required cleaning and have never become blocked.

Ochil View, Lumphinnans

Following the success of the Shettleston scheme, a similar minewater-sourced heat pump system was constructed in 2000 in connection with the refurbishment of a 1950s apartment block, comprising 18 apartments, at Ochil View, Lumphinnans, near Cowdenbeath, Fife [NT 171 928]. The area is underlain by a part of the Fife Coalfield, known locally as the Lochgelly Coalfield, which has had a long history of coal mining. Lumphinnans XI and XII collieries worked from 1896 to 1966 (Fife Mining Heritage Society 2008), with the water discharge from Lumphinnans No. XI pit being reported as at least

Fig. 2. (**a**, **b**) Two views of the Glenalmond Street housing complex, Shettleston, Glasgow. Photograph by D. Banks.
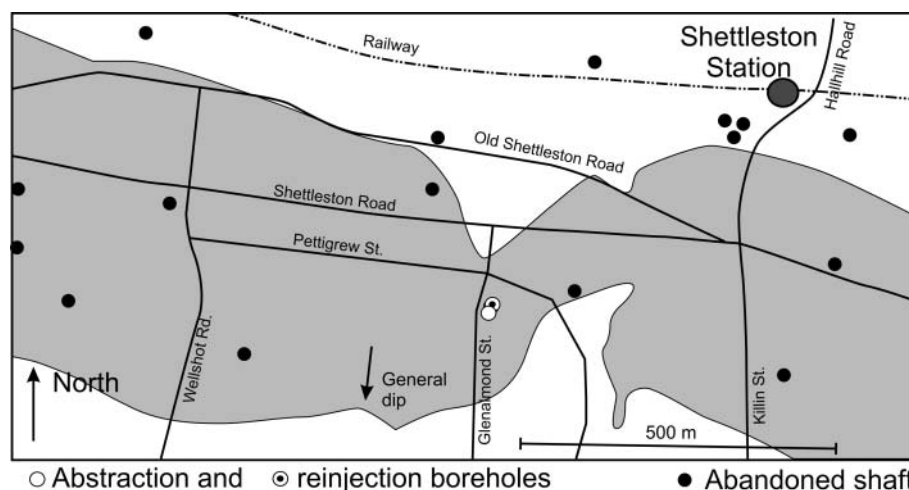

Fig. 3. Map of the Shettleston site, showing the extent of workings in the Ell seam (shaded grey), redrawn from mine plans (especially plan 345) held by the Coal Authority.

$75 \, l \, s^{-1}$ (Burke 1999) prior to closure. The coal-bearing strata belong to the Lower Carboniferous Limestone Coal Group and at least 11 seams have been worked in the vicinity of Ochil View, of which the stratigraphically highest are the Lochgelly Black Band Ironstone, the Seven Foot Seam, the (Kelty) Main, the Jersey or Diamond Seam and the Swallowdrum (Burke 1999).

Minewater is abstracted at a reported temperature of 14.5 °C from workings via a 172.5 m deep borehole, drilled at 8 inches and lined at 6 inches (IPR 1999; John Gilbert 2001). The workings in question are believed to be those of Lumphinnans No. 1 Colliery in the Diamond or Jersey seam, abandoned in around 1944 (Fig. 4). The shallower reinjection borehole is located around 100 m from the abstraction well. As at Shettleston, the minewater is pumped 24 h per day and the water quality and flow are not routinely monitored. A single analysis suggests, however, a pH of around 6.2, an electrical conductivity of 2560 $\mu S \, cm^{-1}$, a reducing Eh and a dissolved iron concentration of 58 $mg \, l^{-1}$ (Table 2).

The combined effect of the building refurbishment and installation of the heat pump system has been to halve the primary energy cost for heating and domestic hot water per apartment and reduce the annual CO_2 emission from 5.2 to 1.9 tonnes per year. Prior to refurbishment the apartments were heated by electric storage heaters (FSHA 2003), using electricity from the national grid.

The Lumphinnans scheme has been more problematic in terms of operation than the Shettleston scheme, although no critical failures can be ascribed to the system concept as such. Indeed, the gauze filters at Lumphinnans generally do not require replacement, and no significant accumulations of ferric oxyhydroxide are observed in the heat pump evaporators or in the associated pipework (S. Johnston, pers. comm.). There have, however, been operational problems related to the heat pumps themselves and, more significantly, in around 2005, vandals damaged the recharge main at the reinjection well, necessitating the well to be redrilled.
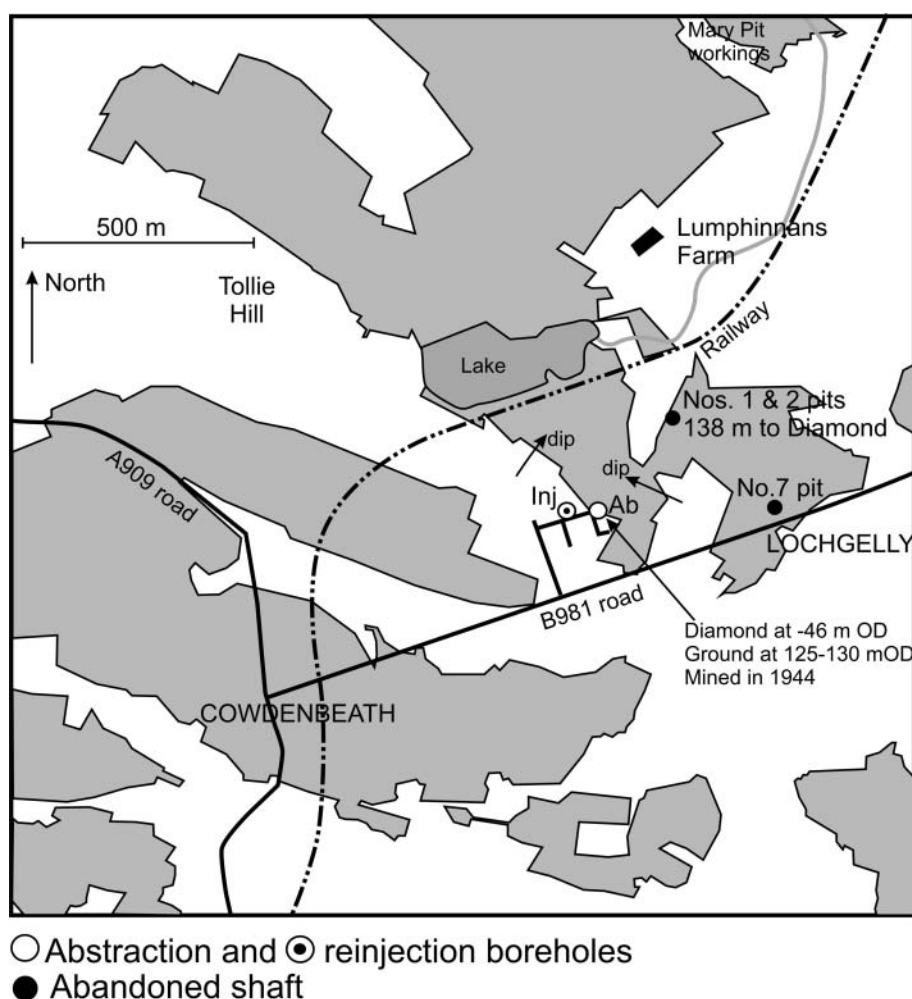
Fig. 4. Map of the Ochil View site, showing the extent of workings in the Jersey–Diamond seam (shaded grey), redrawn from plans held by the Coal Authority (especially plans S4115 5/7 and S406 3/11).

Following redrilling, the recharge water is not 'injected' below the water level in the borehole, but allowed to cascade down the borehole from a manhole (Fig. 5). This has led to precipitation of 'ochre' (confirmed by XRD analysis to contain goethite; Fraga Pumar 2007) in the recharge borehole, clogging of the borehole and periodic overflowing of the borehole at the surface. The submersible pump was found on removal in 2006–2007 to be covered in 'ochre'. The Coal Authority considers a pump lifetime of 5–10 years in ochreous mine water as typical.

The impact of ground source heat extraction on mineral precipitation

The British reluctance to use minewater in open-loop ground source heat systems can, to some extent, be ascribed to the belief that there is a risk of iron and manganese oxyhydroxides precipitating out in heat exchange elements (or in a recharge well) necessitating regular replacement or maintenance. The potential concerns can be summarized as follows.

(1) A temperature change will alter the solubility products and saturation indices of the relevant mineral phases.

(2) Carbon dioxide will degas from the minewater, raising the pH and making oxyhydroxide minerals (or even carbonates, such as calcite) more likely to precipitate. This phenomenon is recognized from minewater treatment (McAllan *et al.* 2009).

(3) Exposure to atmospheric oxygen will oxidize ferrous to ferric iron, in turn causing precipitation of ferric oxyhydroxide minerals.

(4) A change in temperature will increase the rate of oxidation and precipitation of minerals.

(5) An increase in temperature will favour enhanced biofilm formation in heat exchangers or a recharge well.

The first three of these concerns can be investigated using a hydrochemical modelling program, such as PHREEQCI (Parkhurst & Appelo 1999). A sample of water (Table 2), taken from the Lumphinnans discharge main on 25 May 2007 (Fig. 5), was analysed both in the field (for pH, redox potential, temperature, electrical

Table 2. *Minewater sample taken from discharge pipe (Fig. 5) at Ochil View on 25 May 2007*

Parameter	Unit	Filtered sample	Unfiltered sample
Field-determined			
pH		6.16	
Temperature	°C	12.1	
Eh	mV	+29	
EC	µS cm^{-1}	2557	
Alkalinity	mequiv l^{-1}	6.5	
Laboratory-determined			
Chloride	mg l^{-1}	20.3	
Sulphate	mg l^{-1}	1339	
Calcium	mg l^{-1}	315	315
Magnesium	mg l^{-1}	224	225
Sodium	mg l^{-1}	15.6	15.8
Potassium	mg l^{-1}	21.6	21.6
Iron	mg l^{-1}	57.9	58.1
Manganese	mg l^{-1}	3.0	3.0
Aluminium	mg l^{-1}	<0.5	<0.5
Zinc	mg l^{-1}	<0.1	<0.1
Silicon	mg l^{-1}	3.6	3.7
Ion balance error	%	3.6	
Simulated by PHREEQCI			
Fe^{II}	mol l^{-1}	1.0×10^{-3}	
Fe^{III}	mol l^{-1}	7.6×10^{-11}	
CO$_2$	atm	$10^{-0.72}$	
Calcite (CaCO$_3$)	SI	-0.8	
Chalcedony (SiO$_2$)	SI	-0.5	
Dolomite (CaMg(CO$_3$)$_2$)	SI	-1.5	
Fe(OH)$_{3(a)}$	SI	-2.7	
Goethite (FeOOH)	SI	$+2.7$	
Gypsum (CaSO$_4$.2H$_2$O)	SI	-0.3	
Pyrolusite (MnO$_2$)	SI	-22.6	
Rhodochrosite (MnCO$_3$)	SI	-0.3	
Siderite (FeCO$_3$)	SI	$+0.7$	

The heat pumps were not believed to be running at full capacity at the time of sampling, hence the temperature is probably close to undisturbed conditions. The water had an odour of H$_2$S.

Fig. 5. The vandalized discharge main at Lumphinnans. Thermally 'spent' minewater cascades into a manhole; the recharge borehole itself is underwater. Accumulation of 'ochre' can be seen, especially around the mouth of the discharge pipe.

conductivity and alkalinity) and at the laboratory of the Hydrogeochemical Engineering Research and Outreach (HERO) at Newcastle University. The program PHREEQC Interactive version 2.13.2 (Parkhurst & Appelo 1999) has been used to simulate the speciation of inorganic components in the minewater (Table 2), using the PHREEQC thermodynamic database. It is found that the water is in a reducing condition, with iron and manganese overwhelmingly present in the Fe^{II} and Mn^{II} oxidation states. The water has a significant excess partial CO$_2$ pressure of $10^{-0.72}$ atm, compared with around $10^{-1.5}$ atm in many 'normal' groundwaters (reflecting soil CO$_2$ concentrations) and $10^{-3.5}$ atm in the atmosphere. The high partial CO$_2$ pressure suggests that protons generated by pyrite oxidation (equation (2)) have been neutralized by carbonates in the formation (calcite, ankerite, siderite), releasing excess inorganic carbon to the groundwater. Another potential source of excess CO$_2$ may have been sulphate reduction in reducing niches within the mine or aquifer system. The water is undersaturated with respect to ferrihydrite (taken to be Fe(OH)$_{3(a)}$ in the PHREEQC database), pyrolusite (MnO$_2$) and calcite. The water is only slightly undersaturated with respect to rhodochrosite (MnCO$_3$), chalcedony (SiO$_2$) and gypsum (CaSO$_4$.2H$_2$O), and is slightly oversaturated with respect to siderite (FeCO$_3$).

PHREEQCI has been used to simulate the potential for mineral precipitation as a result of change in temperature, degassing of CO$_2$ and contact with atmospheric oxygen. It should be emphasized that PHREEQCI has been applied in this context only to explore thermodynamic tendencies to precipitation or dissolution; no attempt has been made to address the likely importance of kinetic factors or microbiological catalysis of such reactions. When simulating precipitation of iron oxyhydroxides in a heat exchanger or recharge well, one must consider the mineral phase that is likely to be precipitated. As it is generally accepted (see Langmuir 1997) that a semi-amorphous hydroxide (often termed ferrihydrite Fe(OH)$_3$.nH$_2$O) is the normal product of rapid hydrolysis of Fe^{3+} and/or oxidation of Fe^{2+}, amorphous iron hydroxide (assumed to be synonymous with ferrihydrite in PHREEQCI) was selected as the critical phase for the purpose of modelling. Figure 6 illustrates that changing the temperature of the minewater alone results in little change to the calculated saturation indices. Indeed, a temperature change of ±6°C (comparable with the temperature change occurring in most ground source heat pump systems) does not result in either calcite or iron hydroxide becoming oversaturated.

PHREEQC has also been used to simulate the progressive degassing of CO$_2$ in the event of exposure of minewater to the atmosphere or to significant decreases
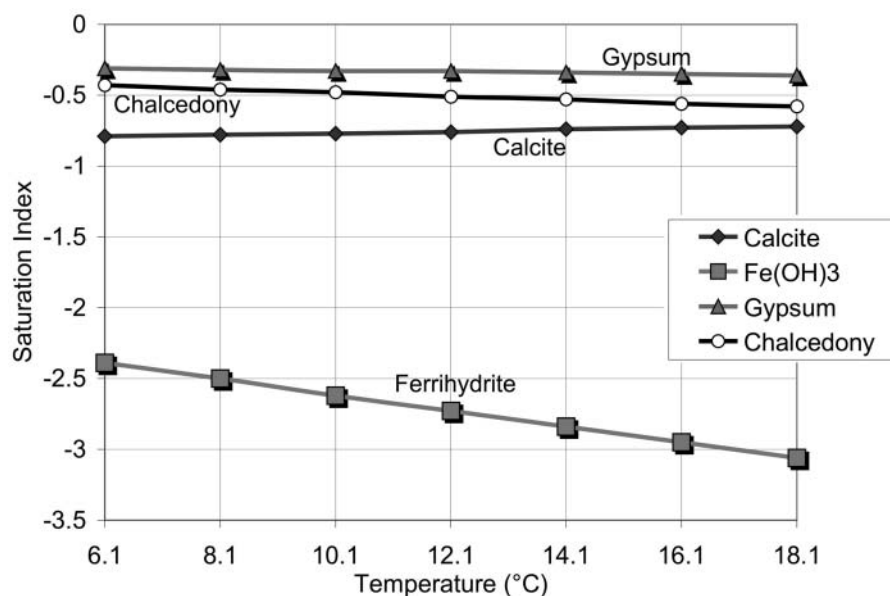
Fig. 6. Modelled saturation indices using PHREEQCI for the Lumphinnans minewater (Table 2), varying only temperature in 2°C increments. The term ferrihydrite is assumed synonymous with amorphous $Fe(OH)_3$ in the context of the PHREEQCI database.
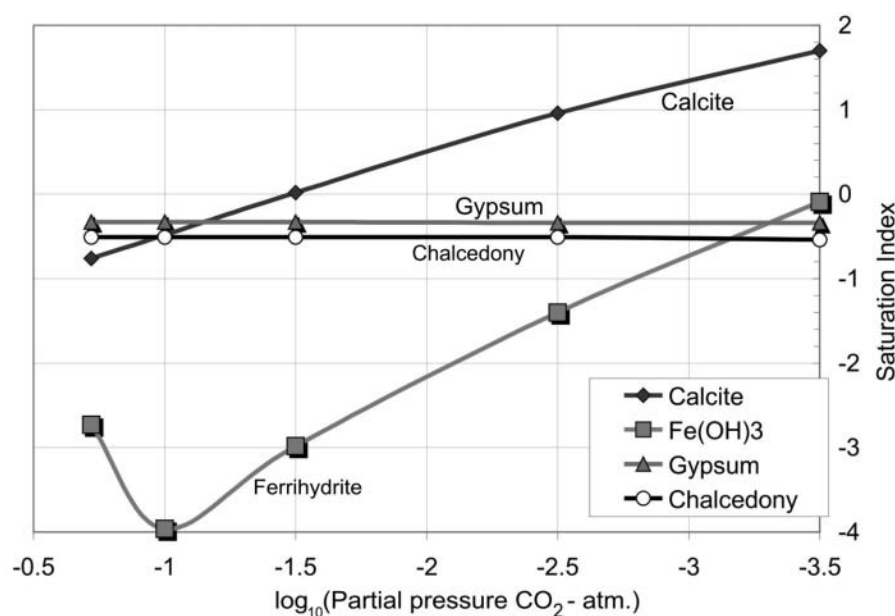

Fig. 7. Modelled saturation indices using PHREEQCI for the Lumphinnans minewater, varying only partial pressure of CO_2. The initial minewater (Table 2) has a partial pressure of CO_2 of $10^{-0.72}$ atm; this is allowed to degas to an atmospheric value of $10^{-3.5}$ atm.

in pressure within the heat pump–recharge system. The Lumphinnans minewater has been progressively equilibrated with CO_2 partial pressures of $10^{-1.0}$, $10^{-1.5}$, $10^{-2.5}$ and $10^{-3.5}$ atm, the last partial pressure being comparable with the atmospheric partial pressure of CO_2. The simulated degassing of CO_2 leads to a significant rise in pH as protons are consumed from solution:

$$HCO_3^- + H^+ = CO_2 + H_2O. \qquad (5)$$
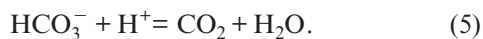
This leads, in turn, to a progressive increase in the calcite saturation index, becoming saturated at $PCO_2 =$

$10^{-1.5}$ atm and significantly oversaturated by $PCO_2 = 10^{-3.5}$ atm. It also leads to an increase in the saturation index of ferric hydroxide, with the solution becoming approximately saturated if an equilibrium with the atmosphere ($PCO_2 = 10^{-3.5}$ atm) is achieved (Fig. 7).

Finally, the exposure to atmospheric oxygen was simulated by progressively adding oxygen to the solution in PHREEQCI (Fig. 8). We can see from the equation for oxidation of ferrous iron
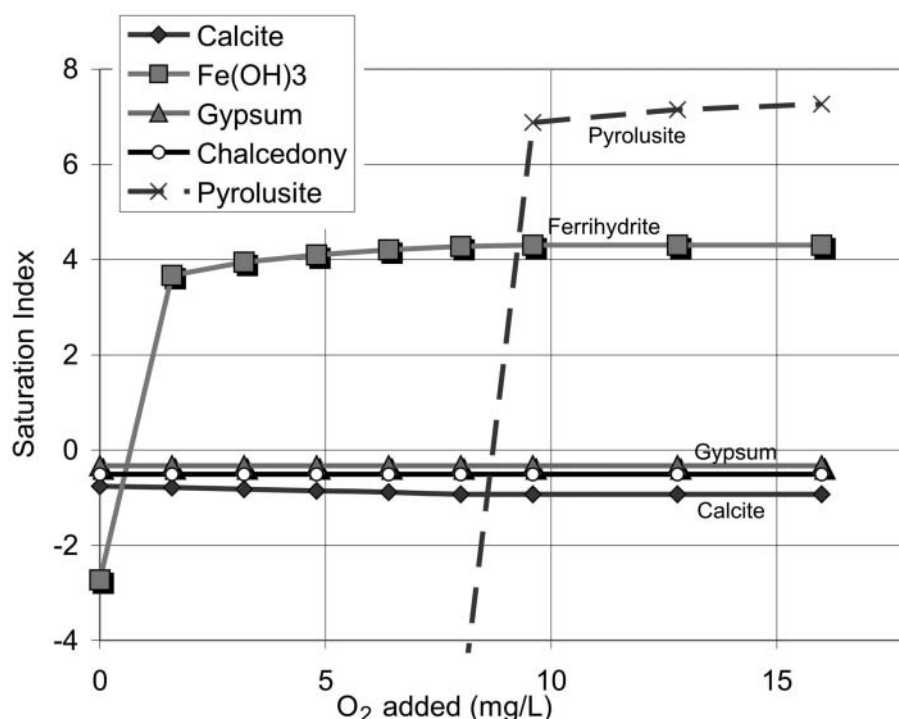
$$4Fe^{2+} + O_2 + 4H^+ = 4Fe^{3+} + 2H_2O \qquad (6)$$

Fig. 8. Modelled saturation indices using PHREEQCI for the Lumphinnans minewater, varying only exposure to O_2. The initial minewater (Table 2) is reacted with O_2 in 0.05 mmol l^{-1} (1.6 mg l^{-1}) increments.

that each mole of ferrous iron requires 0.25 moles of oxygen for complete oxidation. The Lumphinnans minewater contains around 1 mmol l^{-1} of ferrous iron. In theory, therefore, 0.25 mmol l^{-1} oxygen (8 mg l^{-1}) are required to completely oxidize it. We can see from Figure 8, however, that after the addition of only a few mg l^{-1} oxygen, sufficient ferrous iron has been oxidized to the ferric form for ferric hydroxide to become over-saturated and potentially precipitated. As more oxygen is added, the ferric hydroxide saturation index increases further. After 8 mg l^{-1} oxygen has been added, the iron is fully oxidized and the ferric hydroxide saturation index increases no further. At this point, oxygen is consumed in oxidizing manganese (+II) and pyrolusite ($Mn^{IV}O_2$) rapidly becomes saturated.

As regards the kinetics of iron oxidation and precipitation, the following observations can be made. The rate of abiotic oxidation of ferrous iron by oxygen (equation (6)) is known to be first order with respect to iron and oxygen, and second order with respect to OH^-, at pH > 5 (Singer & Stumm 1970; Stumm & Morgan 1996):

$$\frac{-d[Fe^{2+}]}{dt} = kP_{O_2}[Fe^{2+}][OH^-]^2. \qquad (7)$$

At pH < 4, the abiotic rate becomes independent of pH. The abiotic oxidation rate increases about tenfold for every 15°C temperature increase (Sung & Morgan 1980; Stumm & Morgan 1996). At low pH, the presence of iron-oxidizing microbes also becomes critical and can result in several orders of magnitude more rapid oxidation. Biotic oxidation rates decrease with increas-

ing pH, however, and become less significant than abiotic oxidation above pH 5–6. Again, overall (biotic + abiotic) oxidation rates increase significantly with temperature (Kirby *et al.* 1999).

Conclusions

As a result of PHREEQCI modelling and consideration of kinetics, we can reach the following conclusions regarding the tendency of the Lumphinnans minewater to cause chemical clogging in heat exchangers or a recharge well.

- Pressure reductions in the pumping system and/or exposure to atmospheric carbon dioxide concentrations have the potential to result in degassing of carbon dioxide from the minewater. This results in a potential hazard of CO_2 gas accumulation in confined spaces and in a rise in pH of the minewater, leading to a thermodynamic tendency for the precipitation of calcite and iron hydroxide. Of these, ferric hydroxide precipitation would be expected to be the greatest potential hydrochemical problem, as its precipitation is surface catalysed and can be potentially rapid; moreover, it is rather voluminous in its amorphous forms.
- Exposure to only a small amount of atmospheric oxygen can lead to ferric hydroxide (and even pyrolusite) becoming significantly oversaturated and potentially precipitating.
- Net heating schemes, which result in chilling of the abstracted groundwater, would be expected to

result in a reduction in the rate of biotic and abiotic iron oxidation (if thermodynamically favourable) and probably also in the rate of biofilm formation. Net cooling schemes, where heat is dumped to the groundwater flow, would be expected to result in an increase in the rate of iron oxidation and biofilm formation.

- If, however, the pumped minewater is not exposed to atmospheric oxygen or carbon dioxide concentrations, nor to significant changes in pressure, the change in temperature of the minewater alone, as it passes through the heat exchanger, is not sufficient to result in significant changes in saturation indices of potentially precipitating mineral phases, nor in oversaturation of calcite, pyrolusite, chalcedony or ferric hydroxide.

- Thus, if pumping does not result in dewatering of the flooded mine horizon and admixture of oxygen, if the pipework and heat exchanger are sealed units, if recharge takes place well below the water level in the recharge well and if pressures are regulated in the system (e.g. by regulation valves on the discharge main in the recharge well), the potential for precipitation of ferric and manganese oxyhydroxides and of calcite should be minimized.

These conclusions wholly support the operational experiences from Shettleston and Lumphinnans; that is, that significant mineral precipitation has not been observed in the pipework, heat exchanger or recharge wells under normal operation. Following the vandalism of the recharge main at Lumphinnans, however, such that admixture of oxygen has occurred, significant iron oxyhydroxide precipitation has occurred within the recharge well.

Although the findings of this study cannot automatically be extrapolated to other mine systems and water chemistries, they offer grounds for optimism that, by careful management of system pressures and minimizing exposure to atmospheric conditions, minewater-based GSHP schemes can function reliably and with minimal maintenance requirements.

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