The Environmental Legacy of Historic Mining Activities in the Almond River Catchment, Scotland

Simon Haunch[1], Alan MacDonald[2], Neil Brown[3], Christopher McDermott[1]

[1]Edinburgh University, School of Geosciences, Kings Buildings, Edinburgh, EH9 3JW, S.Haunch-2@sms.ed.ac.uk; [2]The British Geological Survey, Kings Buildings, Edinburgh, EH9 3LA; [3]West Lothian Council, County Buildings, High Street, Linlithgow, EH49 7EZ

Abstract The Almond River Catchment, Scotland, has experienced significant coal and oil shale mining over the last 300 years. Examination of surface water quality data from 1994 to 2008 indicates that this legacy continues to impact water quality. During low flows, elevated iron and sulphate loadings in the river are correlated with both coal and oil shale mining areas, most likely from pyrite oxidation at abandoned mine sites. On certain river sections iron loading decreases due to in-stream precipitation. Both iron and sulphate concentrations in the river water show significant flow dependence due to: the re-suspension of river bed iron precipitate; and sulphate dilution.

Key Words- Oil shale, water quality, Scottish mining

Introduction

Production of heavily mineralised discharge waters is a phenomenon observed worldwide at active and abandoned mine sites. Mine closure commonly results in increases in the concentration of dissolved ions in discharge waters caused by groundwater rebound and subsequent flooding of the mine. Discharge of these waters at surface can have serious environmental consequences for the recipient rivers and surface water bodies. Waste rock brought to the surface in the process of mining can also produce similar discharge waters on exposure to atmospheric oxygen and precipitation.

Coal mining is widespread throughout the UK and Europe. The risks to the environment from abandoned coal mining and the mechanisms of contaminant production are, therefore, well studied and characterised (e.g. Younger 2001). Contaminant production in the form of elevated levels of iron (Fe^{2+}, Fe^{3+}) and sulphate (SO_4^{2-}) in mine discharge waters has widely been attributed to the oxidation of pyrite, either by the ingress of oxygen and/or oxygen rich waters into a subsurface mine system or surface mine waste. When these waters discharge at surface iron precipitate ($Fe(OH)_3$) is formed, due to the oxidation of dissolved iron. This results in the characteristic orange ochre seen at mine sites around the world. Proton acidity (H^+) is also produced by pyrite oxidation, however, levels of acidity are usually mitigated in discharge waters by carbonate mineral reactions in the mine, a process termed 'carbonate buffering'. In Scotland where most coals are associated with carbonate rich rocks, such as limestone, discharge waters are, therefore, generally maintained at circum-neutral pH. Mine waste discharge waters, however, usual produce more acidic discharge waters due to lower pyrite-carbonate mineral ratios (Rees *et al.*2002).

The environmental impacts of oil shale mining are poorly characterised compared to other, more common, forms of mining. Oil shale and coal bearing rocks have similar pyrite contents, up to 5%, and both are usually associated with marine limestones. Pyrite oxidation and carbonate buffering reactions are, therefore, likely to be of principle control on the chemistry of oil shale mine discharge waters, as with coal. Oil shale mines in Scotland (Carruthers *et al.*1927) and Estonia (Erg 2005) have been documented as producing contaminated waters similar to those associated with abandoned coal mines.

The Scottish Oil Shale industry exposed mined oil shales to temperatures above 500 °C to extract the organic content of the shale as a form of crude oil (Louw and Addison 1985). Industrial processing of the shale by this method is likely to have oxidised any pyrite in the shale, preventing further pyrite oxidation when the shales are deposited as waste. Scottish oil shale wastes have been noted as containing significant amounts of iron- Fe_2O_3-12% and sulphur- SO_3-3.2% (Burns, 1978). Weathering of the oil shale waste may result in the release of iron and sulphur in discharge waters, however, probably at lower concentrations than those caused by the comparatively vigorous oxidation of pyrite. This paper outlines the scale and distribution of historic oil shale and coal mining activities in the Almond River Catchment and uses surface water quality data to assess environmental impact.

The Study Area

The Almond River Catchment is located in the central belt of Scotland, between Glasgow and Edinburgh (Fig. 1). The area has a history of over 200 years of intensive mining for both coal and oil

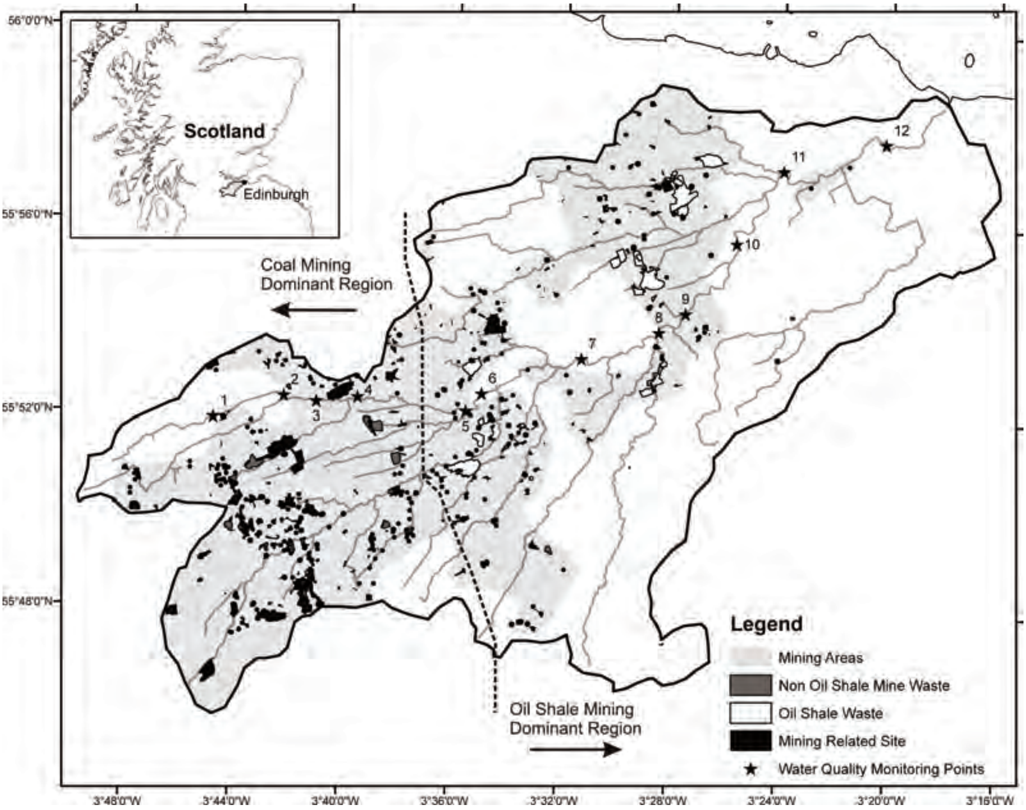

***Figure 1** The Distribution of Mining, Mine Waste and Oil Shale Waste in the Almond River Catchment.*

shale which has resulted in significant impacts on the quality of surface water (Pollard *et al.* 2001). Over 300 sites relate to the extraction or disposal of mined or quarried mineral resources in the catchment, the majority of which being coal and oil shale mines, although less amounts of limestone, slate, sandstone, metals and clay were also mined. Scottish shale oil was produced, almost entirely, within the river catchment due to the mining of oil shale from the Scottish oil shale group.

Methods

Extensive and detailed datasets of the geology/hydrogeology, water quality and land use history of the Almond River catchment were acquired from the British Geological Survey (BGS), Scottish Environment Protection Agency (SEPA) and West Lothian Council (WLC) respectively. These data were used in an integrated catchment scale investigation of the Almond River Catchment.

Spatial Analysis – Mine datasets provided by WLC and the BGS were refined through field observations and analysis of historic data sources (Winter 2001, MacDonald *et al.* 2003). Mine location data was refined sufficiently to be able to confidently identify the dominant mine types (i.e. subsurface, opencast or mine waste site) and mined mineral resources. Historic land use data was then compared, in ARC GIS, to the geological and hydrogeological data. Principle mining areas relating to different mined resource were identified. Spatial comparison of field data and surface water quality data sets was then undertaken.

Water Quality Data - Scottish Environment Protection Agency (SEPA) provided data for all monitoring stations in the catchment for the purpose of this study. Iron and sulphate data were used to assess the impact of mining on the quality of water in the Almond River Catchment. Daily flow readings from four gauging stations in the catchment were used in calculating flow readings at each monitoring station. Flow readings at the monitoring stations were calculated by multiplying the flow reading at the nearest gauging station by a correction factor calculated from the relative catchment area ratio between the monitoring station and the nearest gauging station. This method of flow calculation was deemed suitable as the cal-

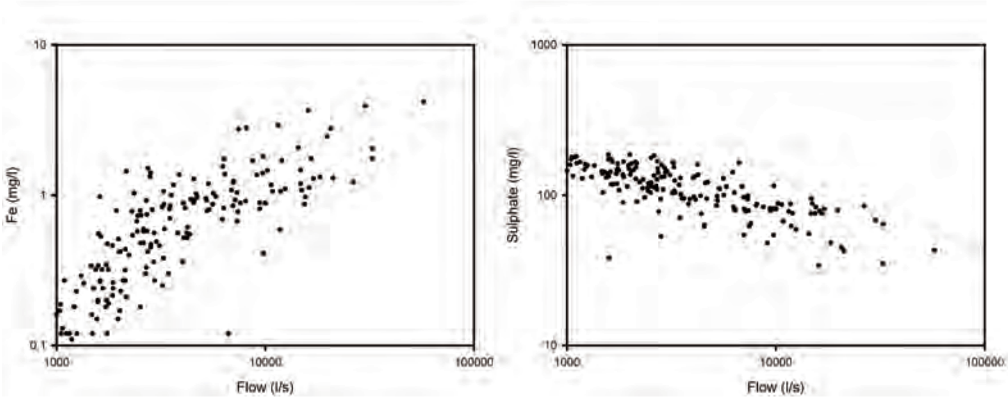

Figure 2- *Concentration flow dependence of iron and sulphate at monitoring point 12 in the Almond River Catchment.*

culated catchment ratios where high (close to 1) due to the proximity of sample stations to gauging stations. Flow readings were then used to either analyse the relationship between river water chemistry and flow or to calculate the loading of the selected chemical parameters across the Almond River catchment.

Mining

Mining in the Almond River catchment dates back to the pre-industrial revolution however it was not until the late 18th, early 19th century that large scale industrial coal mining became widespread. Oil shale mining started later, in the 1860's, with the invention of the oil shale heating and extraction method, developed by James Young. Early shallow mining targeted the most accessible coal and oil shale seams and produced small amounts of mine waste. Advances in technology allowed deeper mining, producing increased mine waste. The total volume of mine waste from the oil shale industry is estimated at 150 million tonnes (McAdam 1993). The volume of coal waste is thought to be much less, particularly, as some of the main coal mine areas of the catchment have since been redeveloped. Coal was transported to be burnt in homes and factories whilst oil shale was processed and then deposited within the catchment in large waste accumulations, known locally as 'Bings'. Almost the entire mined volume of shale oil industry was deposited as waste following the extraction of the oil shales' ≈ 14% organic content. Oil shale mining went into decline in the early 20th century, due to competition from foreign oil, finally closing in the 1960's after the withdrawal of over forty years of government tax relief (Louw and Addison 1985). Coal mining continued until widespread UK closure began in the 1980's. Mine closure in an area with such significant amounts of close proximity mining (Fig.1) has resulted in the continued production of large volumes of mine discharge water following groundwater rebound.

The scale and distribution of mining and the resulting mine waste across the Almond catchment is not uniform and several distinct areas of mining activity can be identified. Coal mining occurred in the south west of the catchment, targeting coals in the Scottish Coal Measures, Passage Group and Limestone Coal Group. Oil shale mining occurred in the central east of the catchment targeting the 20 workable oil shale seems in the Oil shale group. This produces a clear geographic divide between coal and oil shale mining in the catchment. The exact number, distribution and chemistry of mine discharges across the catchment are unknown. Reduced water quality and how it relates to the distribution of mine sites, in Fig.1, is used here to asses the environmental impact of mining in the catchment.

Water Quality

Elevated median concentrations of iron (0.680—1.59 mg/L) and sulphate (91—177mg/L) in waters sampled at 12 monitoring points (Fig.1) on the main tributary of the Almond (Table 1), demonstrate the impact of mine discharges on the quality of water. Comparison of concentration data to the hydrological conditions at the time of sampling reveals that in stream concentrations are often heavily flow dependent. Iron and sulphate concentrations recorded at monitoring point 12, on a monthly basis over a 15 year monitoring period 1994—2008, when compared to flow data show two different flow dependent relationships; iron concentrations increase with increased flow whilst sulphate concentrations reduce with increased flow (Fig.2). All 12 of the monitoring points along the Almond River, although it is less pronounced at monitoring points with small catchment areas, display the same relationships. At monitoring point 12 the relationship of iron to flow can be approximated to a power law with and an $R^2 = 0.635$. Flow dependence of iron in surface waters has been observed, although over shorter monitoring periods with smaller distribution of flow values, in similar heavily mined catchments where it has been related to a change in the source of in-stream iron (Mayes 2008). Dissolved iron, when exposed to oxygen, precipitates rapidly producing iron rich precipitates coating river beds in heavily mined areas. Re-suspension of these precipitates is thought to be responsible for the increases in iron concentrations recorded at high flow values.

Sulphate shows a different flow dependent relationship as it does not readily precipitate on exposure to oxygen and, unlike iron, is not stored on river beds ready to be re-suspended during periods of high flow. This reduction is caused by dilution as the mass of sulphate in the river channel remains near constant whilst the volume of water increases. Iron and sulphate concentrations, therefore, as the main contaminants associated with mining will best represent the impact of mining on water quality at low flow values. At higher flow values the input of iron from mine discharges are masked by the re-suspension of river bed iron precipitates (Mayes 2008). Iron concentrations at high flow maybe useful as an indication of the amount and location of the 'stored' river bed iron.

Concentration data was also used to calculate loading at each of the 12 monitoring points; loading being a measure of the total mass of iron or sulphate that passes each monitoring point. Increases in load between monitoring points represent the input of iron and sulphate from mine discharges, whilst reductions represent precipitation. Figure 3 shows the box plot distribution of loading values, calculated from the concentration and flow data, for iron and sulphate at monitoring points along the main Almond River tributary during low flow events 1994—2008. Low flow is defined here as flow values falling below the 30th percentile for the distribution of flow values recorded at each monitoring point during the monitored period (1994—2008 Fe, 1994—2006 sulphate). The box plots for the load distribution at each monitoring point show 5th, 10th, 25th, 50th (median), 75th, 90th, 95th, percentiles and the mean. Geometric mean is used for the iron loading values due to the power law relationship of iron and flow outlined previously. The mean value of the distribution of loading at each point is joined by the dashed line to give a visual representation of the mean loading profile along the river. An upward trend on the loading profile represents the input of fresh iron or sulphate, whilst a downward trend represents precipitation. The plots show between a 1 and 2 order magnitude increase in loading of both iron and sulphate from one end of the catchment (point 1) to the other (point 12). This is a clear indication of the impact of iron and sulphate heavy mine discharge waters, controlled by pyrite oxidation, on the quality of surface water.

The geographic divide between coal and oil shale mining (Fig.1) in the Almond Catchment allows more detailed interpretation of load increases observed in the loading profile. In addition to the visual assessment of the profile indicating the impact of each area on loading in the Almond river water, cumulative loading can be calculated from the sum of the loading increases along the river to give a semi-quantitative assessment. Sulphate cumulative loading is not calculated due to the lack of data at points 7, 8, 9 and 11.

Coal mining areas show mean low flow iron and sulphate loading increases between monitoring point 1 and 3 and point 5 and 6 indicating the input of iron and sulphate from coal mine discharge waters. The cumulative low flow iron loading increase between points 1 and 6, attributed to coal mining, is calculated at 0.113 g/s. Discharges from both subsurface mining and surface water deposits are likely to contribute to the recorded load increases.

The increase in the iron load, of 0.204 g/s, between monitoring points 6 and 7 is attributed to a mix of coal and oil shale mine discharges as tributaries entering the main Almond river tributary here drain both coal and oil shale mining areas. Sulphate is not monitored at point 7 although a load increase would be expected.

Oil shale mining areas show mean low flow iron load increases between point 7 and point 9 and iron and sulphate load increases between point 10 and point 12 which indicate the input of iron and sulphate from oil shale mine discharges. The cumulative low flow iron loading increase between points 7 and 12 is calculated at 0.115g/s. Discharges from oil shale waste as well as subsurface mine discharges maybe contributing to the recorded load increases. Subsurface mine discharge waters are, however, thought to dominate because the industrial processing of the oil shale is likely to diminish the pyrite content. Weathering of the waste may release iron and sulphate, mentioned previously, which could potentially contribute to observed load increases.

Loading values (Table 1) and profiles (Fig.3) calculated at low flow in the Almond catchment clearly demonstrate that both coal and oil shale mined areas contribute to iron and sulphate loading on the main Almond tributary. Indicating both areas discharge significant volumes of mine discharge water to the surface water environment.

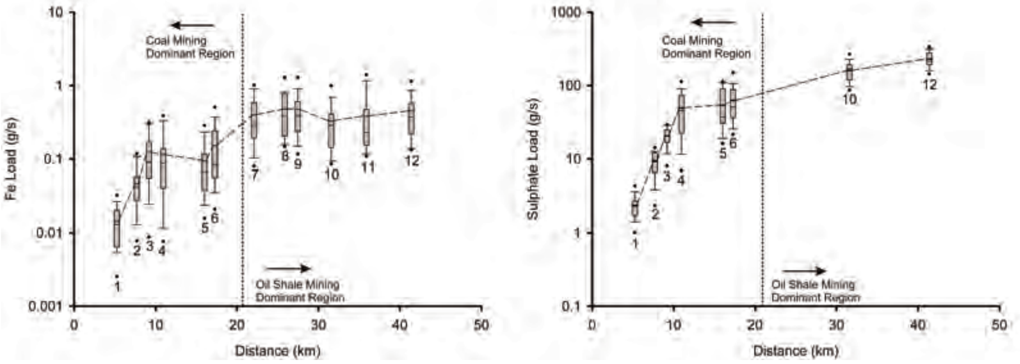

Figure 3 *Box Plot of the distribution of Iron (1994—2008) and Sulphate (1994—2006) loading values at low flow vs. distance*

Table 1 *Iron and Sulphate concentration and loading at 12 monitoring points on the main Almond River Tributary- geometric means are used to for iron due to flow dependence of concentration values.*

Iron	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**	**12**
Median (mg/l)	1.265	1.195	1.590	1.300	0.880	1.175	1.300	1.015	0.915	0.790	0.779	0.680
Mean (mg/l)	2.367	1.599	1.668	1.486	1.075	1.206	1.260	1.134	1.071	0.955	0.928	0.815
Low flow (g/s)	0.012	0.038	0.089	0.072	0.068	0.104	0.308	0.387	0.389	0.256	0.276	0.371
High Flow (g/s)	3.058	4.678	9.445	9.706	10.104	15.378	30.164	50.273	46.620	49.059	50.397	34.739
Sulphate												
Median (mg/l)	91.900	127.500	131.500	178.550	176.000	177.000	n.d.	n.d.	n.d.	124.000	n.d.	114.000
Mean (mg/l)	96.223	132.129	132.447	244.192	230.740	192.438	n.d.	n.d.	n.d.	120.600	n.d.	113.887
Low flow (g/s)	2.376	9.246	20.585	49.416	53.895	62.876	n.d.	n.d.	n.d.	161.190	n.d.	230.283

Cumulative iron loading increases along the length of the monitored section calculated at 0.432 g/s, will in part be derived from natural sources, however, due to the scale of mining in the catchment (Fig.1), mining is though to be the dominant iron source.

Iron load whilst increasing from one end of the catchment to the other also show a reduction in load between monitoring point 3 (0.089g/s), 4 (0.072g/s) and 5 (0.068g/s) and between point 9 (0.389g/s) and 10 (0.256g/s). Precipitation of iron, out of solution, between monitoring points accounts for these reductions in load. Iron precipitates readily in the presence of oxygen, however, a load reduction will only be observed in monitoring data when the mass of precipitated iron is greater than mass of the input of 'fresh' iron i.e. in tributaries which do not receive significant volumes of water from mine discharges. This appears to be the case in the Almond catchment; river sections where a load reduction is recorded generally do not have any close proximity mine sites (Fig. 1). Sulphate loading does not show any reduction in load between 3, 4 and 5 and although sulphate is not monitored at point 9 it is unlikely any reduction would be observed because sulphate precipitation is not oxygen dependent.

During high flow events iron loading is up to 130 times greater than loading at low flow. High flow is defined here as flow values falling above the 70th percentile for the distribution of flow values recorded at each monitoring point during the monitored period. Load is increased to this level due to the flow dependence of iron concentrations (Fig.2) which are increase by the re-suspension of river bed iron. High flow load increases between all monitoring points except point 8 and 9 and point 11 and 12 indicating stored river bed iron is re-suspended from river beds across the catchment.

Conclusions and Further Work

This catchment scale study has highlighted the scale and distribution of coal and oil shale mining in the Almond River catchment and the continued impact that these historic activities have on the quality of surface water. Historic coal and oil shale mining has produced a semi urban landscape with over 200 mine sites and significant volumes of coal mine waste and oil shale waste. Water quality monitoring and flow data show clear contaminant concentration and flow relationships with iron being heavily flow dependent, approximated to a power law relationship. This relationship is attributed to the re-suspension of iron 'stored' on rivers bed across the catchment.

Low flow load reductions in monitoring data demonstrate precipitation and 'storage' of iron on river beds. High flow load increases demonstrate the re-suspension of iron occurring on river beds across the catchment. Low flow iron and sulphate load increases occur in both coal and oil shale dominant mining areas. This indicates that both mine types impact the quality of river water in the catchment. Iron and sulphate levels in discharge waters are controlled, primarily, by the oxidation of pyrite at the mine site. Coal mine waste is thought to contribute to the water quality impacts, however, the contribution of oil shale waste to iron and sulphate loading in oil shale areas is at present difficult to determine. Further investigations into the pyrite content of oil shale waste and the chemistry of discharge waters would be required to confirm and quantify the impact of oil shale waste on water quality.

Acknowledgements

This research is funded through the Edinburgh collaborative of subsurface science and engineering, ECOSSE, programme. The authors thank the Scottish Environment Protection Agency (SEPA) for access to water quality monitoring data for the purpose of this study.

References

Burns J (1978) The Use of Waste and Low-Grade Materials in Road Construction: 6. Spent Oil Shale. TRRL Laboratory Report LR818. Transport Research Laboratory, Crowthome.

Carruthers RG, Caldwell W, Bailey EM, Conacher HRJ (1927) Oil Shale of the Lothians, Memoirs of the Geological Survey, Scotland, Third Edition.

Erg (2005) Changes in Groundwater Sulphate Content in Estonian Oil Shale Mining Area, Oil Shale 22 No.3, 275—289

Louw SJ, Addison J (1985) Studies of the Scottish Oil Shale Industry, Volume1, History and Mineralogy, Institute of Occupational Medicine, Historical Research Report, TM/85/02.

Mac Donald AM, Browne MAE, Smith NA, Colman T and McMillian AA (2003) A GIS of the extent of historical mining activities in Scotland: explanatory notes. British Geological Survey Commissioned report, CR/03/331. 12pp.

Mayes WM, Gozzard E, Potter HAB, Jarvis AP (2008) Quantifying the importance of diffuse minewater pollution in a historically heavily coal mined catchment, Environmental Pollution 151, 165—175

Pollard P, Devlin M, Holloway D (2001) Managing a complex river catchment: a case study on the River Almond, The Science of the Total Environment 265, 343—357

Rees SB, Bowell RJ, Wiseman I (2002) Influence of mine hydrogeology on mine water discharge chemistry, from: Younger PL, Robins NS, Mine water Hydrogeology and Geochemistry, Geological Society, London, Special Publications, 198, 379—390

Winter MG (2001) Spent Oil Shale use in Earthwork Construction, Engineering Geology 60, 285—294.

McAdam AD, Smith RA, Ross DL (1993) Geology for land use planning: Livingston, British Geological Survey, Technical Report WA/92/37.

Younger PL (2001) Mine water pollution in Scotland: nature, extent and preventative strategies, The Science of the Total Environment 265, 309—326.