Basin-scale fluxes of metals from historically mined orefields in the UK

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Abstract. The flux of metals at the tidal limits of major rivers are an important metric of freshwater contaminant transfer to marine habitats, reported in North-East Atlantic bordering countries under the 1992 OSPAR Convention. This paper critically assesses the OSPAR public archive data to estimate the pollution legacy of base metal mining in the major rivers of England and Wales. The data reported for OSPAR are however limited by sample size (12 annual samples to generate a yearly average) and are strongly biased towards low flow sampling, which is likely to considerably underestimate mean total contaminant flux. Alternative methods for assessing long term metal flux are assessed in the River Tyne (2273km^2) which has been subject to extensive historical metal mining in its south-western headwaters, coal mining in eastern lowlands and heavy industry along tidal reaches. Using an extension of the sediment rating curve/flow duration approach, annual fluxes of contaminants are estimated at major flow gauging stations based on a 20 year data series of water quality and stream flow. At the tidal limit of the Tyne, mean annual fluxes of 128 tonnes Zn/year are estimated which represent a 37% increase on mean annual flux data reported for OSPAR over the period 1991-2003. While reasonable relationships can be obtained between flow and contaminant concentration using these long-term datasets (r^2: 0.4 to 0.8) at larger gauging stations, there is still considerable bias towards low flow water quality data, highlighting the importance of high flow water quality monitoring to improve contaminant-flow relations, and thus flux estimates. These data yield useful information for downstream end-users, such as port authorities and riparian developers who dredge and dispose of metal-contaminated sediments at considerable cost.

Key Words: metal mine, mine water, contaminant transport, flux

INTRODUCTION

Drainage from areas of former metal mining activity can cause persistent instream metal contamination (e.g. cadmium (Cd), copper (Cu), lead (Pb) and zinc (Zn)) over large reaches. This mining-derived pollution occurs through a range of ongoing contaminant inputs from point mine water discharges (e.g. from mine drainage levels and adits) as well as diffuse mining-related sources. Diffuse inputs include runoff from waste rock heaps and processing sites, direct discharge of contaminated groundwaters into surface waters via the hyporheic zone (Gandy *et al.*, 2007) as well as the remobilisation of secondary sources of contamination such as metal-laden fluvial sediments (Hudson-Edwards *et* al, 2008). Such metal mining pollutant sources have been shown to be of long-term detriment to the ecological quality of both riverine (Armitage *et* al., 2007) and downstream estuarine habitats (Giusti, 2001). In north east Atlantic bordering countries, contaminant mass flux data (i.e. flow multiplied by concentration) from tidal limits of major rivers are routinely collected under the obligations of the 1992 Oslo-Paris (OSPAR) Convention which provides a mechanism for fifteen governments to cooperate, monitor pollution from land, and protect the surrounding marine environment. The UK government is a signatory to the OSPAR Convention and as such collects and reports these annual mass loadings for a suite of inorganic and organic contaminants on all major UK rivers. Some of the patterns in these data and their scientific value have been critically appraised by various researchers previously (Littlewood, 1995; Webb *et al*., 1997). However, the potential of the OSPAR data for assessing the legacy of contaminant flux from abandoned base metal orefields in the UK has not been addressed in any great detail. This paper provides a review of recent OSPAR data in England and Wales with specific reference to metals commonly associated with historic metal mining activity: Cd, Cu, Pb and Zn. Preliminary attempts are made to distinguish the river basins where long-term elevated contaminant flux and yield are likely to be predominantly mining-related. The OSPAR data are then examined in the historically mined River Tyne catchment in north east England against alternative measures for quantifying metal flux.

METHODS

OSPAR data

In England and Wales, the OSPAR monitoring puts particular burden on the statutory environmental regulators, the Environment Agency (EA) given the relatively high number of river systems which drain to the sea compared to mainland Europe which is characterised by far fewer higher order stream systems such as the Rhine / Meuse and the Elbe. Over the period 1991-2003, from which data are considered here, loadings data from the tidal limits of 157 rivers were reported. In addition, a large number of additional consented discharges to the tidal reaches of streams are monitored but these are not considered here. Flux is typically interpolated using equation 1.

$$L = K\left(\frac{Ci(QiA)}{n}\right) \tag{1}$$

where: L = total annual mass load; K = time correction factor to account for period of record; Ci = instantaneous contaminant total concentration; Qi = instantaneous flow at time of sampling; A = area adjustment factor where the gauging station lies upstream of the true tidal limit; n = number of samples.

The mean of 12 flux values derived from instantaneous flow and water quality measurements (metal contaminant in the total fraction) are adjusted by a time conversion factor to interpolate an annual mass flux. The data presented below represent the mean of these annual flux values. Contaminant yields below are determined through the division of the flux estimates by river basin drainage area to give values in kg/yr/km^2 which offers a useful indication of the severity of the pollution across the different scales of basin for which data are collected (range between 15km^2 and 9895km^2).

Identifying former metal mining areas

Preliminary interrogation of the OSPAR data was undertaken to assess where former metal mining activity is likely to have been a significant contributor to the measured tidal contaminant flux in the monitored basins. For this purpose a series of spatial analyses were undertaken using the software ArcGIS® v9.2. Delineation and calculation of drainage areas to each of the OSPAR reporting stations was undertaken through processing the 50m Digital Terrain Model (DTM) of England and Wales using standard procedures for filling anomalous sinks in the DTM then using the 'filled' DTM for flow path analysis and the derivation of a raster drainage network. Thereafter two spatial screening exercises were undertaken to identify (1) basins in which there is evidence of historic base metal mining and (2) the urban land cover in each of the basins. The latter provides a coarse indication of the presence of other major sources of contaminant metals associated with industry, urban / highways runoff and sewage discharges. A spatial join was carried out to highlight the presence (if any) of abandoned metal mine sites (from a national dataset of >4000 abandoned metal mine sites collated by the EA) falling within each drainage basin as well as the percentage of each basin which had urban land use (from national land use datasets). A four category classification for the basins was then determined. What is hereafter referred to as a “mining area” indicates the presence of abandoned metal mine sites in the basin and urban land use is <5% of the basin land area (i.e. these are predominantly rural catchments where sources of the contaminant metals assessed are most likely to be related to mineralization and mining activity). A “mining / urban” catchment covers basins which have abandoned mines but also a significant urban cover (>5%). “Non-mining urban” areas are basins with no reported abandoned mines and >5% urban cover, while remaining unclassified basins are presented as “non-mining rural”. This 5% cut-off for delineation of urban and rural is largely arbitrary, being set after examination of the distribution of urban land cover in the 157 basins. The data were examined to ensure that all the basins containing major urban centres (and particularly the (post)-industrial centres of central and northern England) that are known to contain metal polluted rivers due to urban / industrial activity fell within the “urban” categories. All these highly urbanized areas such as Manchester (draining to the River Mersey), Birmingham (River Trent and to a lesser degree the River Severn), Leeds (River Aire) and Sheffield (River Don) have urban land cover in excess of 15% so a precautionary 5% threshold was thereafter set. These four categories provide a coarse indication of where it may be expected that metal contaminant sources are predominantly mining-related (“mining areas”), those with multifarious sources (“mining/urban”), basins in which urban activity is likely to be the predominant source of metals (“non-mining urban”) and “non-mining rural” catchments where metal flux/yield would be anticipated to be lowest.

Contaminant flux estimates in the Tyne

In the Tyne basin, which drains one of the most productive parts of the North Pennine Orefield, a twenty year database of water quality and flow records are used to extrapolate flux values through the formulation of Zn concentration rating curves. Many of the contaminants associated with metal mine pollution are known to exhibit fairly simple flow-concentration responses (e.g. Webb *et al.*, 1997). This is a feature of most metal contaminants being present predominantly in particulate form in river systems and suspended sediment concentration being strongly related to flow condition. The generic rating equation (2) between C (Zn concentration) and Q (flow) (with a and b constants in the power law relation) was applied to generate such relations at 5 gauging stations in the Tyne basin. Four of these lie on the Tyne itself, the most downstream station Bywell (from which OSPAR data are derived), the Haydon Bridge and Reaverhill stations which lie upstream of the confluence of the South and North Tyne respectively and at Alston in the headwaters of the South Tyne in the Pb-Zn orefield. The most downstream station on the River Derwent, a tidal tributary of the Tyne, is also assessed as it drains part of the orefield. These Zn concentration rating curves are used in conjunction with flow-duration curves at each of the stations to assess the long term flux at each of the stations (following an extension of the method for sediment flux calculation detailed by Julien, 1988), while the inter-annual variability in flux at the Bywell station is derived from the rating curve extrapolation and 15 minute flow records and compared with the reported OSPAR data.

$$C = aQ^b \tag{2}$$

RESULTS & DISCUSSION

OSPAR data – the national picture

Figures 1a-d show the mean annual flux over the period 1991-2003 (in graduated circles) and yield (i.e. flux/drainage area, on the shaded colour ramp) for Zn, Pb, Cu and Cd for all reported OSPAR basins. Figure 2a-d meanwhile show the flux data against drainage area with one of the four mining / land use classes assigned to each basin (see legend). In general the higher fluxes of all metals are present in the larger drainage streams as would be expected given the simple scale-dependency of sediment flux (and thus associated metals). However there are a large number of basins where the contaminant yield (i.e. flux per unit drainage area) is far higher than the norm. For Zn, elevated fluxes are particularly apparent in the rivers Ystwyth and Rheidol in Western Wales, the Wear and Tyne draining the North Pennine Pb-Zn Orefield, the River Ouse draining part of the Yorkshire Pennine Orefield, and Restronguet Creek in Cornwall. Of notable interest, the Zn flux of 21 tonnes/year from the Afon Goch (draining the Cu mine complex of Mynydd Parys on the Isle of Anglesey) to the Irish Sea (drainage area $34km^2$), which is of a similar magnitude to that draining the River Mersey (40 tonnes/year, drainage area 3030 km^2) containing much of the conurbation of Manchester. The Zn yield in the Afon Goch is 0.74 tonnes $Zn/yr/km^2$ which is second only to the Restronguet Creek (South West) which is a massive 1.9 tonnes $Zn/yr/km^2$ given the proximity of the mine site to coastal discharge in this small catchment. Large fluxes of all metals are apparent in the large basins of the Trent, Severn and Thames, with the peak recorded Zn flux on the River Trent of around 145 tonnes/year. While the former two drain the orefields of the South Pennines and West Shropshire respectively, they are also likely to receive substantial metal flux from urban areas. This is particularly the case for the Trent which drains ~470 abandoned metal mining sites in the South Pennines, but also the West Midlands to south of the Trent catchment which was a major focal point for the automobile industry in the mid to late 20^{th} Century. One other notable non-mining outlier in the Zn flux data is the River Tawe in south Wales, which while not containing any abandoned metal mine sites and being predominantly rural, was a major centre for refining of Zn, Pb, Cu and Ni in the Lower Swansea Valley for over 250 years until the 1980s.

Catchments draining the Pennine Pb-Zn orefields are prominent sources of Pb, particularly those draining east into the Humber (Ouse, Wharfe, Aire and Don) and west into the Ribble and Mersey. Of these, the Ouse and Wharfe are predominantly rural (urban cover of 3% each) and the sources of the Pb are well-documented in both the wider Ouse basin (Hudson-Edwards *et al.*, 1999) and the sub-catchment of the River Swale in particular (e.g. Coulthard & Macklin, 2003) to be associated with the gradual dispersion of mining-related sediments through the river systems. The Aire, Don and Mersey have large urban cover (29%, 31% and 20% respectively) and were former centres of heavy industry which are likely to account for the significant portion of the contaminant flux despite the presence of some former metal mine sites in these basins. The major north eastern rivers of the Tyne, Wear and Tees are major contributors of Pb to the North Sea. While heavy industry has dominated the tidal reaches of these rivers, the OSPAR reporting stations lie upstream and the predominant metal mining related sources in these basins have been well-characterized (e.g. Lord and Morgan, 1996; Nuttall and Younger, 1999). The Rivers Rheidol, Ystwyth and Dyfi draining the rural western Wales orefield and the River Dee, which drains both the Halkyn Mountain / Minera orefield of north eastern Wales and large industrial districts in lower reaches, also report high Pb flux and yields (maximum of 47 kg $Pb/yr/km^2$ in the Dyfi).

The largely urban catchments of the Thames, Trent, Mersey, Severn and Aire are major sources of Cu, with the peak recorded flux again in the River Trent (33 tonnes/yr). Mining-related sources are apparent in the Restronguet Creek and River Tamar in Cornwall, and in the Afon Goch which drains the Mynydd Parys mine complex, all of which show high Cu flux in the region of 10-20 tonnes/year. The Cu yields from the Afon Goch and Restronguet Creek, which are both small coastal catchments containing severely polluting mine sites are

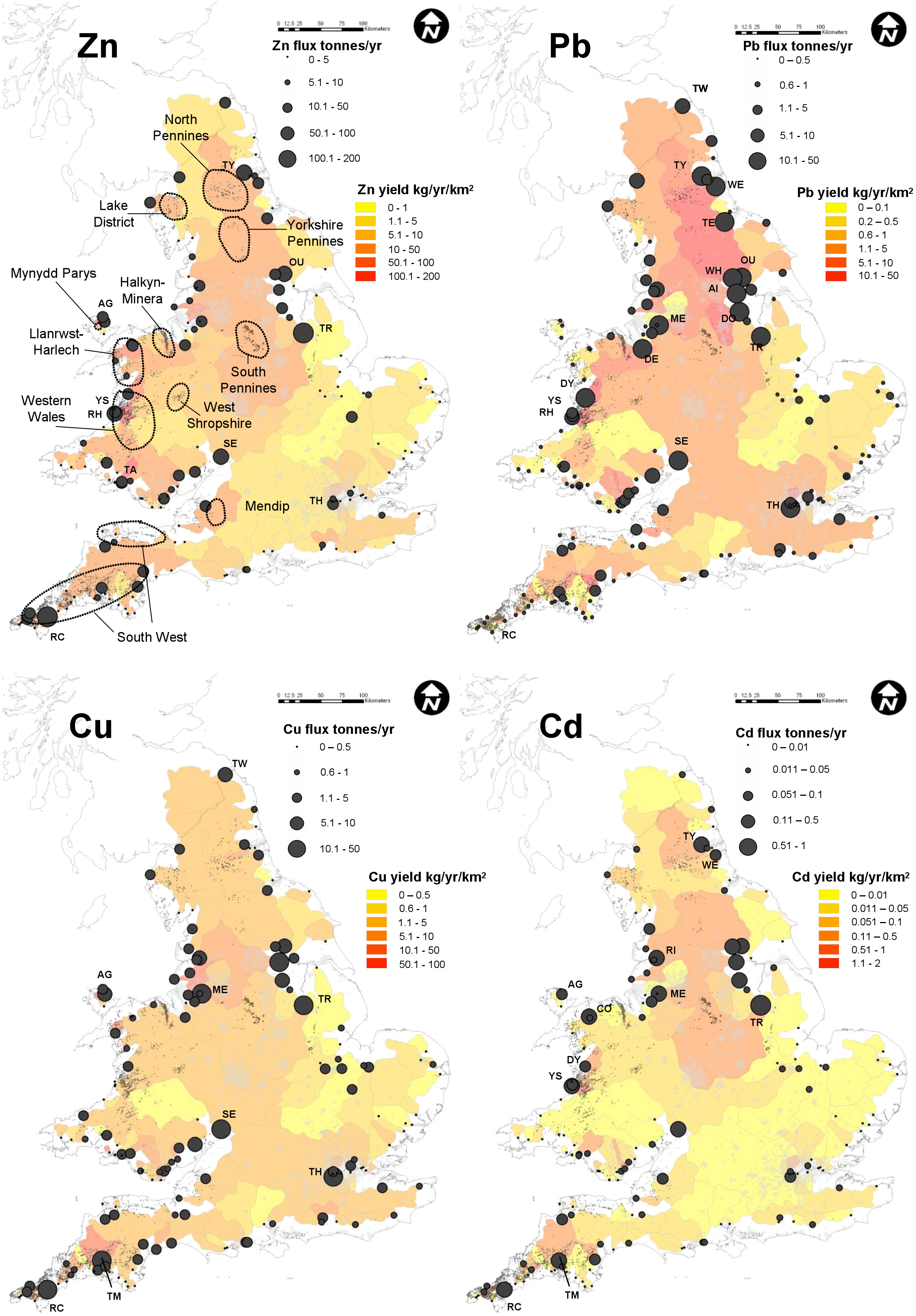
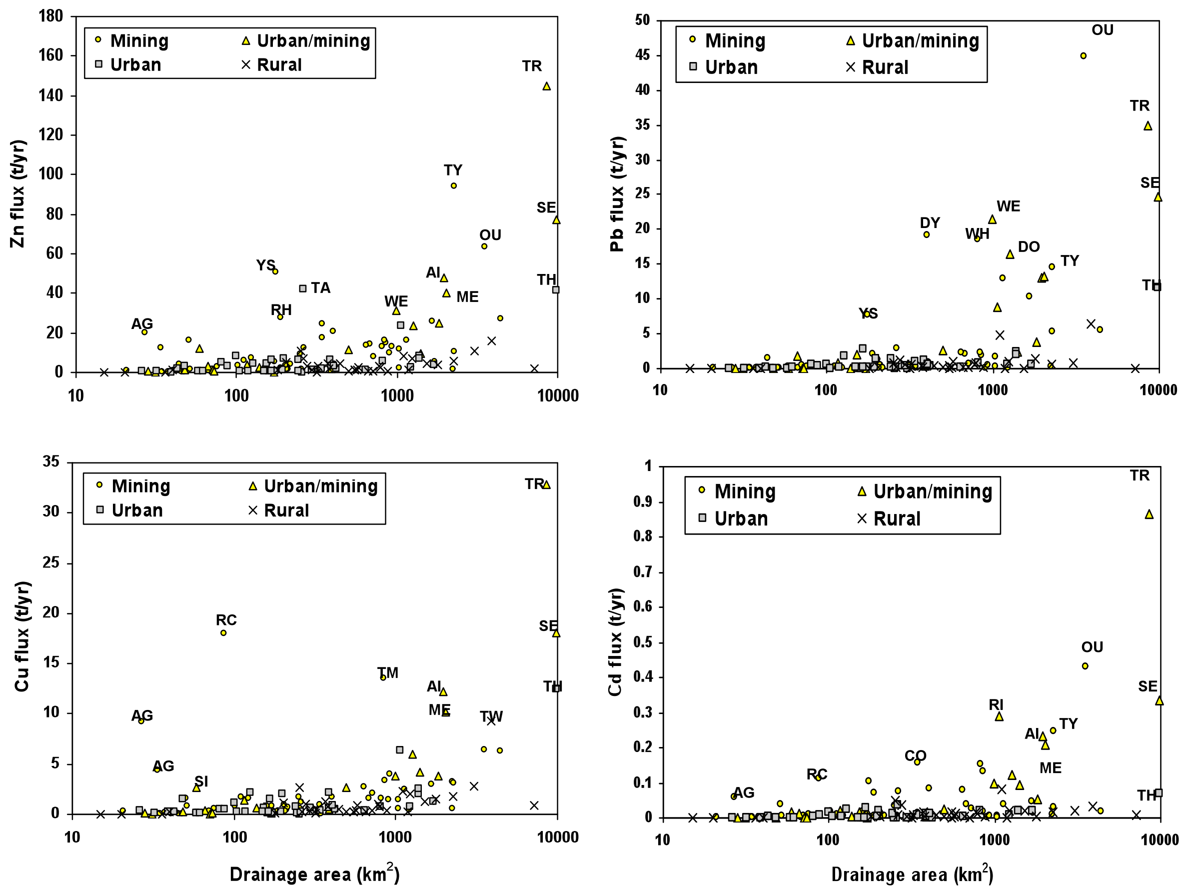


Figure 1. Mean annual OSPAR flux and yield data for four metals (1991-2003). Small black dots indicate former mine sites. Main orefields annotated on Fig 1a only. Urban areas



Selected annotated stations: AG = Afon Goch, Anglesey (Western Wales River Basin District, note: there are two reporting stations on the Afon Goch); AI = River Aire (Humber); CO = River Conwy (W. Wales); DO = River Don (Humber); DY = River Dyfi (W. Wales); ME = River Mersey (North West); OU = River Ouse (Humber); SE = River Severn (Severn); SI = St. Ives Bay (South West); RC = Restronguet Creek at Deveron Bridge (South West) RH = River Rheidol (Western Wales); RI = Ribble (North West); TA = River Tawe (W. Wales); TH = River Thames (Thames); TM = River Tamar (South West); TR = River Trent (Humber); TW = River Tweed (Northumbria); TY = River Tyne at Wylam (Northumbria); WE = River Wear (Northumbria); WH = River Wharfe (Humber); YS = River Ystwyth (W.Wales).

Figure 2. Mean annual contaminant flux (1991-2003) reported under OSPAR against drainage area for four river basin classes.

both in excess of 200 kg Cu/yr/km^2. One slightly anomalous station in the Cu data is the River Tweed, the northern most basin monitored in eastern England, where a relatively high mean flux of 9.3 tonnes/year is reported. This is likely due to drainage from a small, but dense cluster of electrical manufacturing industry in Selkirk in the central Tweed basin (Robson & Neal, 1997).

With the exception of the Trent, Mersey, Aire and Severn, the higher fluxes of Cd are associated with drainage from areas where pollutant sources are likely to be predominantly mining-related. The Pennine orefield rivers (Tyne, Wear, Ouse, Wharfe), the major rivers of western Wales (Ystwyth, Dyfi and Rheidol), the Conwy and Afon Goch (draining the Llanrwst-Harlech and Mynydd Parys orefields respectively), and the Tamar and Restronguet Creek (south west England) all show high yields of Cd as illustrated in their elevation above other data points in Figure 2d. Peak Cd yields are again reported in the Restronguet Creek (1.3 kg Cd/yr/km^2) and Afon Goch (2.2 kg Cd/yr/km^2). For comparison the Cd yield in the Trent is 0.1 kg Cd/yr/km^2.

The total mean contaminant flux from all monitoring stations aggregated into the four broad basin classifications is presented in Table 1. This highlights that approximately half of the Zn and Pb flux is associated with basins where there is a fair degree of confidence that contaminant sources are not related to contemporary industrial / urban activities. Slightly less than 40% of the Cu flux is associated with mining areas, while 45% of the Cd reported to discharge to coastal waters of England and Wales drains base metal orefields.

Table 1. The total mean annual flux (tonnes/yr) of metals reported at the tidal limits of major rivers of England and Wales under OSPAR, 1991-2003 for the four basin categories detailed above.

	N	Total area (km^2)	Zn	Pb	Cu	Cd
Non-mining rural area	34	33042	119.0	24.3	36.4	0.4
Non-mining urban area	39	22850	189.6	32.9	40.0	0.4
Mining / urban area	21	29691	443.9	138.0	101.1	2.1
Mining area	58	35978	817.9	170.1	115.6	2.4
Total	152	121561	1570.4	365.3	293.1	5.3

While these data give a coarse indication of the large contribution made by basins in which former mining activity has taken place, there are of course other potentially significant sources of contaminants within these mining areas. These could include natural weathering of metal-rich minerals and the release of metals deposited by atmospheric fallout (e.g. Rothwell *et al.*, 2007). One such example of this non-mining related enrichment could be the Ribble catchment in north-west England which reports relatively high yields of Cd despite the former metal mining activity being known to be of minimal impact on water quality at a local scale. However, in most of the basins consistently reporting both elevated fluxes and yields of the contaminants considered here, more focused mine site and occasionally catchment-scale studies have recognized the mining activity as the principal agent of instream and sediment metal enrichment (see Hudson-Edwards *et al.,* 2008).

Metal flux estimates – a case study from the Tyne catchment, north east England

The River Tyne is a major source of Cd, Pb and Zn flux to the North Sea. These metals are predominantly related to sources in the South Tyne sub-catchment draining part of the North Pennine Orefield which was worked extensively for Pb, Zn and fluorspar amongst other minerals up until the mid 20th century. Flow-Zn concentration relations (Eq. 2) were formulated for 5 gauging stations in the basin based on twenty year water quality data and flow records (Fig. 3, Table 2). While the power law function yielded reasonable relationships at two of the stations, 2 stations were better described by linear relationships. Other workers have also found that flow-sediment and flow-contaminant curves in some basins are better described by linear or polynomial relationships (e.g. Webb *et al.*, 1997). At the most upstream station (Alston), no significant relationship could be formulated due to dispersion in the dataset. If anything the relation at Alston appears to show a very weak negative correlation from high ambient baseflow concentrations (due to ongoing mine water inputs) which are diluted at higher flow. However in smaller basins the effects of occasional peaks associated with flushing episodes, diurnal variability in concentration and differences in summer and winter baseflow concentrations are likely to impart far greater variability in the concentration data than in larger basins where baseflow concentrations are far lower (due to dilution) and effects of flushing are dampened. Table 2 shows the relationships for the curves and the resultant long-term flux estimates when the relations are applied to flow duration curves at each of the gauging stations. The mean total flux at Bywell is 123 tonnes/yr. When this value is adjusted for drainage area (in the same manner in which the OSPAR data for the Tyne catchment is reported) then a long-term mean flux of 128 tonnes/yr is estimated, 37% higher than the mean flux reported for OSPAR over 1991-2003. The data also reveal the very significant contribution to the downstream flux made by the South Tyne sub-catchment as would be anticipated given the location of the former metal mining area. The Derwent station shows a moderate flux of Zn of 3.8 tonnes/yr which is 10% higher than the mean OSPAR value over the period 1991-2003, the latter of which is considerably elevated by a value of 12 tonnes/yr reported in 2003. While mining-related sources are likely to contribute to the contaminant flux in the Derwent there is also a large urban coverage in the lowlands of the catchment (21% urban land cover) and former industrial sites (e.g. steelworks) that could contribute to the reported downstream values.

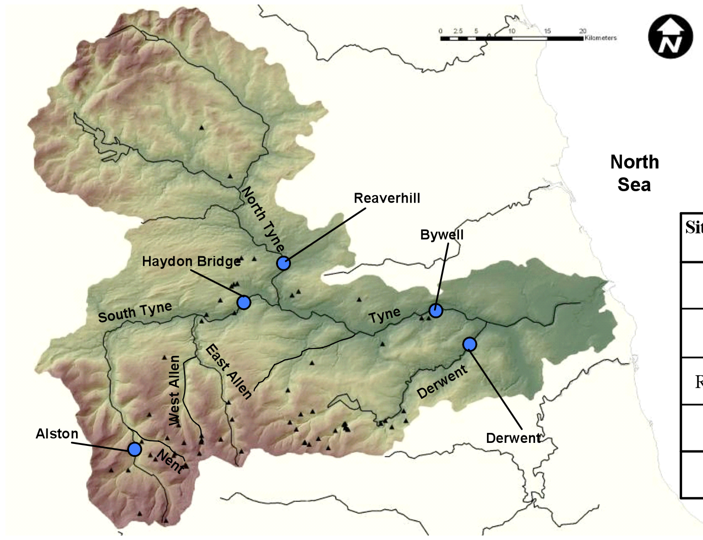


Table 2

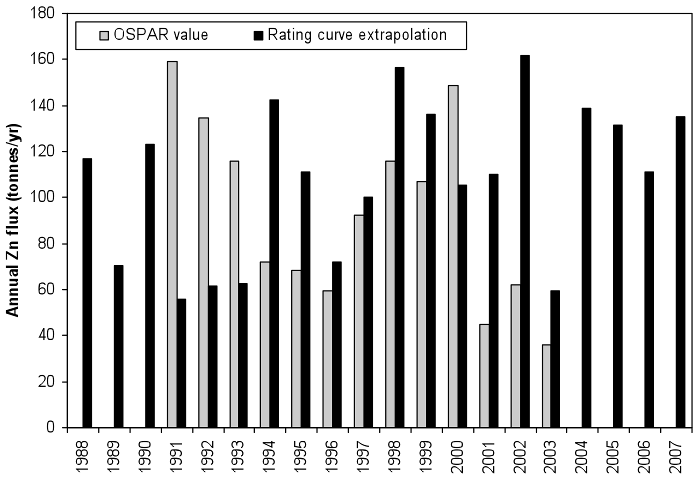
Site (drainage area km²)	Expression	R^2	Flux
Alston (119)	$C = 57.5Q^{-0.0281}$	0.047	-
Haydon Bridge (751)	$C = 1.9Q + 82.0$	0.802*	115.7
Reaverhill (1007)	$C = 0.088Q + 6.4$	0.416*	7.8
Bywell (2176)	$C = 11.3Q^{0.4201}$	0.462*	122.8
Derwent (242)	$C = 5.3Q + 14.3$	0.703*	3.8

*significant at $p = 0.05$

Figure 3. Location map of the Tyne catchment; Table 2. Formulated relations between total Zn concentration and flow and resultant flux estimates based on flow duration curves.

An annual time series of Zn flux from the Bywell station was generated from 15 minute flow records and the relations in Table 2 for the period 1988-2007 and is shown in Figure 4 alongside the reported OSPAR data (1991-2003). The data reveal the strong inter-annual variability in flux estimates from both datasets, with the OSPAR values differing from the extrapolated estimates between 34% and 260%. In general, the OSPAR data report lower figures than the extrapolation with the exception of relatively dry years of 1991-1993 and 2000 which was a very high flow year. As would be expected, the extrapolated values correlate better with cumulative annual flow (r^2 of 0.67) than the OSPAR data (r^2 of 0.19) which is a likely feature of the stochastic vagaries of using instantaneous flow and concentration data from 12 annual samples.

Figure 4. Annual Zn flux in the Tyne catchment reported under OSPAR and through application of the flow-Zn rating curve to a 15 minute flow data set



While the rating curve relationships developed in the Tyne catchment can provide a simple long-term estimate which takes into account all of the water quality data collected over a long sample period (and therefore increase coverage of a range of flow conditions from which to formulate contaminant-flow relations), there are still significant uncertainties in the values these estimates provide. These principally relate to the sparse coverage of water quality data at moderate to high flow. This is illustrated by the fact that at all of the 5 stations 90% of the water quality data were collected at Q90 or above, i.e. the low flows that persist for over 90% of the time. The few individual samples collected at moderate to high flow have an extremely high leverage on the position of the upper end of the rating curve and underscores the need to improve flux estimates through obtaining more water quality samples over the full range of flow conditions.

CONCLUSIONS AND FUTURE RESEARCH

The data collected under the OSPAR convention provide a valuable tool for assessing the spatial distribution of metal contaminant flux across England and Wales. Using a simple GIS-screening based on former mining activity and urban cover the significant flux of Zn, Pb, Cu and Cd that emanates from the historically worked orefields of England and Wales is highlighted. While there are very large uncertainties associated with the OSPAR flux estimates, due to limited annual sample sets that are unlikely to be representative of a full range of flow-contaminant concentration conditions, the consistent application of the reporting procedure nationally allows useful assessment of patterns of potential ongoing impacts on downstream estuarine and coastal waters. An assessment of the OSPAR Zn flux data in the Tyne catchment emphasize the substantial bias towards water quality sampling in low flow. Simple flow-concentration curves were derived for 5 gauging stations in the Tyne basins, and while significant correlation coefficients can be generated for the larger streams, the small number of water quality measurements at moderate-high flow exert a high leverage on the form of the curve. Future monitoring efforts should focus on event-based sampling to characterize the high flow contaminant behaviour and permit the formulation of more detailed contaminant rating curves which can then be used to extrapolate flux and potentially generate more complex algorithms that can incorporate other factors such as flushing episodes and diurnal variability in flux estimates. Further long-term flux assessments at additional nested stations within the Tyne basin (e.g. Featherstone on the South Tyne, East and West Allen, Nent) could be useful in highlighting the specific source areas. These data will be of importance for assessing metal contaminant impacts on estuarine and coastal ecosystems as well as for the management of metal-rich sediments in mining-impacted river basins. While the data here provide a preliminary overview of contaminant flux from areas of former metal mining in England and Wales, more detailed sub-basin scale assessments of the legacy of pollution from abandoned non-coal mines in England and Wales are currently being undertaken and due to be reported later this year (see Jarvis *et al.*, 2008). This will provide a foundation for a remedial programme addressing the most severely polluted metal mine-impacted catchments in England and Wales.

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