The Contribution of Major Historical Orefields to Coastal Trace Metal(loid) Fluxes in North-East England

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Abstract

Mine waters and tailings are key inputs to riverine metal(loid) transport to the coast. However, coastal areas also face local contaminant pressures, e.g. active industries and abandoned landfill sites. Annual riverine metal(loid) flux data were assessed alongside respective fluxes from coastal industries and legacy landfills along the North-East England coastline to quantify relative contaminant contribution. The majority of each contaminant flux (56-97%) entered the sea via riverine transport from inland orefields. However, coastal landfills contributed non-trivial fluxes of some contaminants (<93 t/yr Pb, <38 t/yr Ni, <17 t/yr As), indicating that inland management alone may not sufficiently remediate coastal waters.

Keywords: Coastal Erosion, Legacy Waste, Abandoned Landfill, Contaminant Flux, Source Apportionment, Mining Waste.

Introduction

The environment of the United Kingdom (UK) contains an abundance of mining wastes deposited prior to contemporary waste management practices (Johnston *et al*., 2008). Such 'legacy' mining wastes are distributed predominantly in former orefields, with recent estimates suggesting 52800 ha of mine spoil deposited in the UK (Riley *et al*., 2021). Of particular concern with legacy mine deposits is the potential release of metal(loid) contaminants (e.g. Zn, Pb, As) due to erosion and leaching (e.g. Singer *et al*., 2020), which alongside direct mine water discharges, are known to enter coastal waters via riverine transport (Mayes *et al*., 2013).

In addition to inland deposits, mining wastes are also commonly encountered in coastal areas (covering ≈4500 ha), with coal spoils being the third most prevalent waste type deposited in these regions (Riley *et al*., 2022). Such coastal deposits are also subject to atmospheric weathering and associated contaminant release, but coastal erosion and tidal flooding, which are both predicted to increase in severity and frequency with continued climate change (Toimil *et al*., 2020; Vitousek *et al*., 2017), pose additional risks at these sites (Riley *et al*., 2022). Further to mine-related sources, it is important to consider sources of contamination to the coastal environment that are not mining-related but may release a similar suite of metal(loid)s. Examples of which are legacy metallurgical and industrial waste deposits (covering ≈1000 and ≈3500 ha of coastal land in the UK, respectively; Riley *et al*., 2022) and long sea outfalls from contemporary industrial activity.

Given that multiple sources of potential contaminants enter coastal waters (i.e., transported from inland or originating within the coastal zone itself), it is critical to properly understand the relative importance of each source in order to inform proper environmental management and protect coastal water quality. Contaminant flux assessments are an effective tool in apportioning the relative contribution of these different sources. Indeed, previous national assessments of contaminant flux from abandoned mines in England and Wales (e.g. Mayes *et al.*, 2010; 2013) have been invaluable for understanding the scope of impacts from legacy mine wastes. Therefore, this paper will assess and incorporate the relative diffuse metal(loid) fluxes from coastal mine spoil deposits, in addition to other coastal legacy waste deposits, and contemporary riverine and coastal point sources. This will provide detailed information on contaminant sources at local and regional scales to aid in management decision-making.

Methods

Geographical extent

The coastline of North-East England, between the Rivers Coquet and Tees was used as the geographical extent of this assessment. This coastline was selected as its rivers not only drain the Pb-Zn orefield of the North Pennines and the Great Northern Coalfield, but is also an industrial and populated coastline with a typical distribution of coastal waste deposits (Riley *et al.*, 2022). Two flux assessments were performed for the area, the first being an assessment of riverine flux from inland sources and the second an assessment of flux from coastal sources, as detailed below.

Riverine metal(loid) flux

The riverine flux of select metal(loid)s are routinely monitored at the tidal limits of major rivers in the UK by the Environment Agency as part of the OSPAR Commission framework. As such, three years of flux data were retrieved and summarised to calculate the average annual flux of Zn, Cu, Pb, Cd, Cr, Ni, and As at the tidal extents of the Rivers Coquet, Wansbeck, Tyne, Wear, and Tees (ODIMS, 2023). Note that data for the River Blyth were not available for this analysis.

Coastal metal(loid) flux

For the purpose of this research, two sources of potential contamination were considered to represent the coastal sources of metal(loids); the point-source flux from coastally-located active industries, and the diffuse inputs from eroding coastal legacy landfill sites. Point source inputs were also retrieved from the Environment Agency OSPAR data for the aforementioned metal(loid)s of interest. These sites represented 28 individual point inputs within the region of interest, which were a mix of sewage treatment work effluents and industrial outfalls. Flux data were again averaged to calculate average annual flux for each contaminant of interest.

Coastal legacy landfill sites (identified as containing either MSW, coal mine spoil, or iron and steel slag) within 250 m of the coastline were extracted from the spatial databases generated by Riley *et al.* (2020, 2021, 2022), and used to calculate diffuse inputs via two processes: release of contaminated sediments by coastal erosion, and metal(loid) release through leachate formation.

To estimate the annual release of sediments by erosion, 5 m resolution LiDAR raster data (dating from 2019 and 2009) were used to indicate the position of the coastline at each point in time (Digimap, 2023). Within ArcMap 10.8 GIS software, these data were then clipped to the spatial extent of each legacy landfill. Using the surface volume tool (as per Riley *et al.*, 2020), the volume of each landfill site was calculated for each time point – the difference between which being an estimate of the volume of material eroded over ten years. This value was divided by 10 to approximate annual material flux in m^3 per year. To convert this to mass, reported bulk density values of 312 and 1,687 kg/m^3 were used for municipal solid waste (MSW) and coal spoil (the two waste types within eroding landfills in this region), respectively (Palanivel and Sulaiman, 2014; Fityus *et al.*, 2008). Unpublished data of mean elemental composition (collected from a range of coastal landfill sites around the UK) were then used to estimate the annual flux of specific contaminants via coastal erosion.

To calculate the potential volume of landfill leachate formed, the annual average rainfall (mm converted to m) across the study area (Met Office HadUK-Grid; Hollis *et al.*, 2022) was first calculated. This was multiplied by the known area of each landfill (m^2, from Riley *et al.*, 2022) to give a theoretical maximum leachate volume (m^3), and then multiplied by a runoff coefficient of 0.5 to represent the portion of rainfall which typically infiltrates landfill soils (e.g. Zafar and Alappat, 2011). This analysis was repeated for each of the 189 landfill deposits in the study region. Unpublished data of leaching rates of As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn from samples of these wastes under freshwater and seawater conditions (generated using standard method BS EN 12457-2; NBS, 2002) were used in conjunction with the estimated leachate volumes to model leachate composition and the annual load of each metal(loid) released from each legacy landfill site.

Results and Discussion

In addition to the five major rivers entering the North Sea in the study region, a total of 10 industrial point discharges were recorded and 190 legacy landfill sites (within 250 m of the coastline). The total landfill area covered 1,664 ha, of which 1,109 ha consisted of MSW, 449 ha of coal mining spoil, and 106 ha of iron and steel slag. Of the legacy landfill sites, 20 were determined to be eroding, of which 7 contained coal spoil and the remainder MSW.

Modelled annual flux estimates indicated that substantial total loads of trace metal(loids) entered the North Sea in this region from the combination of these sources (Table 1). For the majority of metal(loids), riverine transport was the dominant contributor to the coastal zone, though the physical release of contaminated sediments by coastal erosion was also a high contributor. High fluxes were attributed to coastal coil spoil deposits, the leachates from which contributed far higher loads than leachates from other waste types. Interestingly, the total flux of Pb and Zn, commonly associated with mining wastes, was higher from eroded MSW (<22,108 and <51,347 kg/yr, respectively), though this is likely due to the higher number of eroding MSW deposits in the region.

When considering the relative flux of specific elements, it becomes possible to infer key sources and pathways to coastal waters, and the geographical regions in which higher fluxes were observed. Of particular concern due to its toxicity, it was determined that the majority of As flux to coastal waters originated within the coastal zone (11,495 kg/yr) rather than riverine transport from inland

***Table 1** Modelled annual high/low estimates of metal(loid) flux (kg/yr) from riverine sources, coastal point sources, and coastal legacy waste sites (via leachate formation and erosion)*

Source		As	Cd	Cr	Cu	Ni	Pb	Zn
Riverine Transport	Low	1,702	507	7,458	23,212	6,741	66,324	192,653
	High	5,016	602	7,929	23,214	23,093	66,354	193,164
Coastal Point Sources	Low	42.0	14.0	610	4,289	370	1,892	10,371
	High	121	29	625	4,289	729	1,893	10,374
MSW Leachate	Low	14.3	1.4	17.9	46.6	17.9	3.6	151
	High	14.3	1.6	28.7	50.2	28.7	7.2	215
Coal Spoil Leachate	Low	0.2	0.1	0.1	0.3	0.5	0.1	7.3
	High	3,743	63.5	1,037	2,043	8,031	387	12,971
Iron & Steel Slag Leachate	Low	0.2	0.0	0.2	0.2	0.2	1.7	0.3
	High	0.2	0.0	1.0	3.4	0.7	1.7	15.7
MSW Erosion	Low	205	4.0	694	363	435	533	2,112
	High	2,000	411	2,594	14,720	3,203	22,108	51,347
Coal Spoil Erosion	Low	594	0.0	869	866	495	332	948
	High	5,616	54	3,012	5,562	2,770	2,747	13,799
Total	Low	2,558	527	9,649	28,777	8,060	69,086	206,243
	High	16,511	1,161	15,227	49,882	37,855	93,498	281,886

sources (5,016 kg/yr; Fig. 1). Approximately 31% of this coastal flux (3,640 kg/yr) was via the formation of landfill leachates, which represent a diffuse aqueous source. Geographically, the more dominant fluxes of As were located between the Tyne and Tees, with a typically higher flux from all sources between these rivers. The area between the Wear and Tees was a particular hotspot for As release via coastal erosion (1,446 kg/yr), likely due to the prominence of eroding coal spoil deposits in this region (representing 6 of the 9 eroding landfills on that stretch of coastline).

Contrary to As, the majority of modelled Ni flux was attributed to riverine sources (23,093 kg/yr vs. 11,059 kg/yr), with the River Tyne being the highest single contributor (9,615 kg/yr; Fig. 2). Nevertheless, coastal sources still contributed a non-trivial flux of Ni to the environment (11,059 kg/yr), the majority of which via the formation of aqueous leachates, albeit in a diffuse manner across multiple legacy waste sites. Whilst minor in comparison, a reasonable flux of Ni from contemporary point discharges (<729 kg/yr) was observed (Fig. 2).

The relative flux assessment of Pb (Fig. 3) revealed strikingly different patterns than As or Ni, with the vast majority (85%) of the total flux originating from riverine sources. Coastal Pb flux was dominated by the erosion of contaminated sediments with a relatively low contribution from landfill leachates. Given the low solubility of Pb in solution (Gao and Bradshaw, 1995), it is perhaps unsurprising that modelled leachates contributed little towards Pb flux. However, despite its low solubility, the high flux of Pb from aqueous sources (especially rivers) illustrates the widespread scale of the issue of Pb transport from historical orefields and the potential impacts this may be having in coastal regions.

Conclusions

It is apparent that substantial fluxes of potentially problematic metal(loids) are entering coastal waters along the North-East coast of England, but it is only through relative flux assessments, such as this, where the relative contributions of different sources may be determined. Through consideration of coastal sources of metal(loid) flux, such

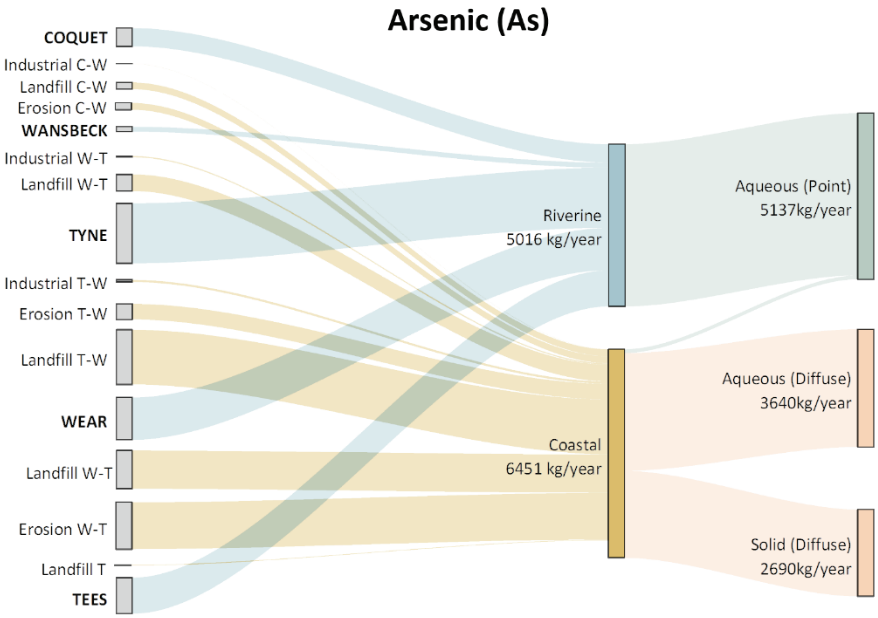

Figure 1 *Relative annual flux of As (kg/yr) from riverine and coastal sources ("industrial" = industrial discharges, "landfill" = landfill leachates, "erosion" = eroded material, suffixes relate to the source position between rivers, e.g. "C-W" = Coquet – Wansbeck). Major rivers are listed from North to South (top to bottom).*

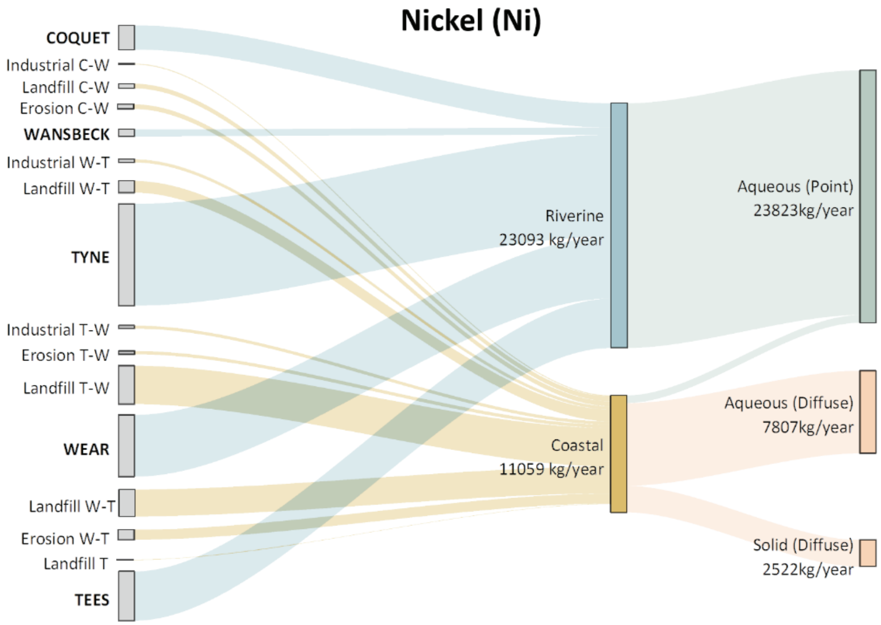

Figure 2 Relative annual *flux of Ni (kg/yr) from riverine and coastal sources. Major rivers are listed from North to South (top to bottom).*

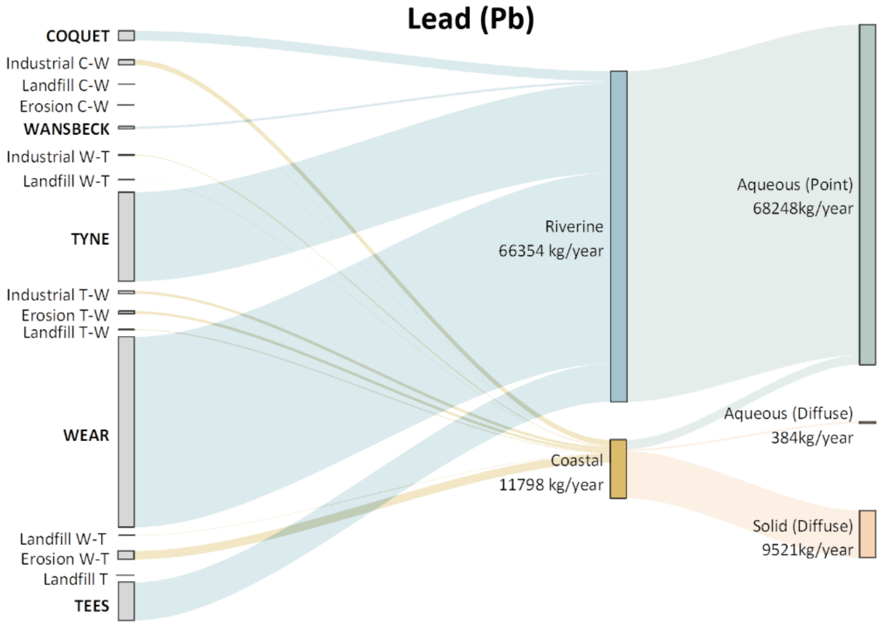

Figure 3 Relative annual *flux of Pb (kg/yr) from riverine and coastal sources. Major rivers are listed from North to South (top to bottom).*

as contemporary industrial discharges and legacy waste landfills (including mining wastes), alongside traditional riverine flux assessments, it is possible to determine the contribution of mining wastes to coastal fluxes in a holistic manner.

The influence of major historical orefields to coastal metal(loid) fluxes (via riverine transport) varied depending upon the specific metal(loid) in question. For some, e.g. Pb and Ni, it was determined that inland sources were likely the main contributor to coastal fluxes, whereas for As, the contribution of coastal sources was more prominent. Further to this, the source and transportation pathways of contaminants (i.e. point or diffuse, aqueous or solid) were assessed, indicating that while aqueous point sources (e.g. rivers, industrial discharges) are major contributors, diffuse aqueous and solid sources (i.e. landfill leachates and eroded material) should not be overlooked. Findings such as these are critical for ensuring good coastal water quality, as effective management will depend on appropriately addressing the exact sources of specific contaminants. Whilst contemporary point discharges may be easier to control, diffuse contamination from coastal landfills and riverine transport of contaminants from historical orefields may require landscape-scale management, which brings additional challenges.

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