



## Research article

## Evidence-based logic chains demonstrate multiple impacts of trace metals on ecosystem services

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## ARTICLE INFO

## Keywords:

Copper  
Mercury  
Bioaccumulation  
Ecosystem good  
Ecosystem process  
Impact chains

## ABSTRACT

Trace metals can have far-reaching ecosystem impacts. In this study, we develop consistent and evidence-based logic chains to demonstrate the wider effects of trace metal contamination on a suite of ecosystem services. They demonstrate knock-on effects from an initial receptor that is sensitive to metal toxicity, along a cascade of impact, to final ecosystem services via alterations to multiple ecosystem processes. We developed logic chains to highlight two aspects of metal toxicity: for impacts of copper pollution in soil ecosystems, and for impacts of mercury in freshwaters. Each link of the chains is supported by published evidence, with an indication of the strength of the supporting science. Copper pollution to soils (134 unique chains) showed a complex network of pathways originating from direct effects on a range of invertebrate and microbial taxa and plants. In contrast, mercury pollution on freshwaters (63 unique chains) shows pathways that broadly follow the food web of this habitat, reflecting the potential for mercury bioaccumulation. Despite different pathways, there is considerable overlap in the final ecosystem services impacted by both of these metals and in both ecosystems. These included reduced human-use impacts (food, fishing), reduced human non-use impacts (amenity value) and positive or negative alterations to climate regulation (impacts on carbon sequestration). Other final ecosystem goods impacted include reduced crop production, animal production, flood regulation, drinking water quality and soil purification. Taking an ecosystem services approach demonstrates that consideration of only the direct effects of metal contamination of soils and water will considerably underestimate the total impacts of these pollutants. Construction of logic chains, evidenced by published literature, allows a robust assessment of potential impacts indicating primary, secondary and tertiary effects.

## 1. Introduction

## 1.1. Trace metals in the environment

Trace metals are a vital component in the infrastructure of modern life. They are essential to a wide range of industrial processes and consumer products that deliver societal and economic benefit. However, as the production and use of metals has grown over time, so have the problems and costs associated with the release of metals into the environment (Jiao et al., 2012). Unlike most organic chemicals, metals are not broken down in surface waters, sediments or soil. As a consequence, metal contamination can only be removed by the relatively slow processes of physical removal, cropping, leaching or evasion/volatilisation in the case of mercury (Hall et al., 2006). The long residence times mean that metals can have prolonged impacts on species and ecological communities (McGrath et al., 1995) and

consequently long-term impacts on ecological functions and ecosystem services. These wider effects of trace metals on ecosystem structure and function need to be understood to allow the development of regulatory policies that balance the benefits arising from the production and use of trace metals with the costs associated with environmental impacts on a broad suite of ecosystem services and effects on human health and wellbeing.

## 1.2. Sources of metals to the environment

The major sources of trace metal releases into air, water and soil include natural sources such as geogenic weathering of parent rocks, volcanism and human activity. Point sources arising from key anthropogenic sources include ore mining, metal processing, industrial production and energy generation. Diffuse sources of human inputs include agricultural uses such as sewage sludge for fertiliser and as biocides

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(McGrath and Cegarra, 1992), copper salts as fungicides (Komarek et al., 2010), aerial deposition from burning of coal and oil for domestic heating (Selin, 2009), and emissions from consumer products including via domestic wastewaters and from product use (Tchounwou et al., 2012). Thus metal contamination can be both highly localised and highly dispersed. With this combination of gross and diffuse inputs, trace elements are some of the most widespread and potentially highest-risk contaminants in soils and freshwaters (Donnachie et al., 2014).

### 1.3. Mechanisms of impact

While releases of trace metals to the environment may occur through similar pathways, their effects on exposed organisms will be highly dependent on the specific toxicokinetic and toxicodynamic interactions of each metal with each organism. Some metals, such as copper, iron, selenium and zinc are essential for the growth of many organisms at low doses but are toxic at high concentrations (Hopkin, 1989). Toxicity effects arise through multiple mechanisms including DNA damage, redox cycling, metabolic toxicity and enzyme cofactor substitution (Nordberg et al., 2014). Other metals, such as cadmium, lead and mercury, are almost exclusively non-essential and, hence, only toxic. Known human health effects resulting from exposure to these metals include hepatotoxicity for cadmium (Ellis et al., 2012; Järup et al., 1998) and neurotoxicity for lead and mercury (Cecil et al., 2008). The molecular mechanisms underpinning these biological effects have non-specific modes of action, such as binding to protein thiol groups, enzyme co-factor substitution and reactive oxygen species production in sensitive tissues. To prevent such toxic effects, some organisms have biochemical systems such as metal chaperoning and storage pathways, to achieve tight control of metal concentrations inside cells (Sakar, 1999).

Differences in metal handling and sensitivity of toxicologically relevant pathways among organisms can result in large variations in species vulnerability to metal exposure. Significant sub-lethal effects can occur at lower exposure concentrations than acute effects, especially when linked to the inhibition of key biological pathways. For example, some key bacterially-mediated ecosystem processes, such as nitrogen fixation, are known to be particularly sensitive to metal (e.g. zinc) exposure (Chen et al., 2015; Judy et al., 2015). Similarly, the effects of metal exposure on metabolic processes can result in reduced feeding activity by soil macroinvertebrates (notably earthworms and Collembola) leading in turn to reduced litter breakdown (Spurgeon et al., 2005). As well as direct effects of exposure, metal pollution can also have indirect effects on associated species. For example, accumulation in tissues of plants and detritivore species can result in the extensive exposure of predators further up the food web. This biomagnification is well documented for metals such as cadmium and mercury (Mason et al., 2000; Spurgeon and Hopkin, 1996b). Additionally, when exposure affects key taxa at a site, then a range of secondary community and ecosystem effects may occur. These interactions and dependencies among organisms, and the ecosystem functions they support, are much less well understood than are direct toxic effects. Such complexity makes the overall consequences of metal pollution for ecosystems more challenging to estimate. Therefore, there is a need for an overarching theoretical framework within which to collate and interpret evidence regarding both primary and secondary effects of metals, in order to fully understand how contamination by trace metals affects ecosystem services.

### 1.4. Use of ecosystem services frameworks to assess wider impacts of pollution impacts

Ecosystem services frameworks are increasingly being used to assess the wider benefits and costs of environmental perturbations, manipulations and management actions. They provide a means to account for

the externalities not usually considered in decision making (Boyd and Banzhaf, 2007; Maltby et al., 2018). Ecosystem services frameworks have already been applied to evaluate the effects of other pollutants, such as nitrogen and ozone, on multiple ecosystem services (Jones et al., 2014; Compton et al., 2012; Hayes et al., 2016). Within these frameworks, logic chains, also called causal impact chains, are being used as a key approach to identify the links between environmental pressures, changes in ecosystem properties (e.g. effects on vulnerable taxa) and effects on ecosystem functions and associated services. For example, Clark et al. (2017) presented a causal chain analysis summarising evidence to link atmospheric nitrogen pollution to impacts on northern spotted owls, via effects on horse-hair lichens and northern flying squirrels. A number of studies are starting to apply an ecosystem services framework to evaluation of trace metal impacts. Some have listed species-specific impacts (Blouin et al., 2013) or have described impacts at the level of service from a remediation perspective (Ding et al., 2018). Approaches that detail the pathways of impact and demonstrate a mechanistic understanding of the effects, evidenced by the literature, bring increased rigour to these assessments and are more transferable to new situations.

Here we use logic chains for the first time to collate and assess the effects of metal pollution on ecosystem services. We refer to these as 'logic chains' as these pathways were derived from a conceptual understanding of potential effects, subsequently verified and evidenced from the literature. We developed logic chains linking exposure to resulting primary, secondary and final effects for copper, an essential metal known to be directly toxic to many organisms at high concentrations, and mercury, a non-essential metal that is both toxic and potentially bioaccumulative. The developed chains consider both similar and unique pathways for these metals within two different ecosystems. For copper, we considered impacts on terrestrial ecosystems, which could potentially result from prolonged application as a fungicide, or as pollution from a smelter. For mercury, we considered exposure of aquatic species from diffuse inputs, including the potential for transfer through the aquatic food chain.

Development of the two sets of logic chains allowed us to identify which ecosystem services are potentially impacted by metal contamination within each scenario, and to highlight knowledge gaps in stepwise logic chains that link direct impacts of the metals on biota to the final ecosystem goods/services affected. Such results are informative for current risk assessment for metals undertaken worldwide (Van Sprang et al., 2004; Hope et al., 2012), and demonstrate how this approach may be applied to other metals.

## 2. Methods

The known impacts of copper and mercury on biota were incorporated into an ecosystem services framework to identify their wider impacts on a suite of provisioning, regulating and cultural services. Primary impacts based on biota were identified from the extensive published ecotoxicological literature available for each metal. Logic chains were developed to link these direct impacts, via subsequent secondary interactions, to impacts on processes or ecosystem attributes. Final ecosystem services dependent on these ecosystem processes and properties were then identified. A schematic illustration of the development of impact chains is shown in Fig. 1. Chains were compared to identify gaps and to ensure that appropriate cross-linkages were in place. Categorisation of final ecosystem services (Provisioning, Regulating and Cultural services) was according to the Final Ecosystem Goods and Services Classification System (Landers and Nahlik, 2013), with a view to recognising the beneficiaries (i.e. users) of the service. Thus cultural services are mainly condensed to human non-use effects, subdivided to amenity, recreation, aesthetic appreciation, education etc. as appropriate. Direct impact on human health was not considered as an ecosystem service endpoint in this study. The end-point of the chain was at the final ecosystem good and was considered in the form

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