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A review of recent advances and future challenges in freshwater salinization

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ABSTRACT

A review of recent advances and future challenges in freshwater salinization

In spite of being a worldwide phenomenon that can have important ecological, economic and social consequences, freshwater salinization (i.e. the increase in ion concentrations in freshwaters) has been poorly studied when compared with other environmental issues. However, it is receiving increasing attention, with significant scientific advances being made during the last two decades. Here I review the current knowledge on the topic and propose future research directions within the context of human welfare and global change. Freshwater salinization is caused by a wide range of human activities, with agriculture and resource extraction being the most widely documented. Different studies suggest that it could be affecting around 1/3 of freshwater bodies, and this number will very likely increase in the future due to climate and land use changes. The increase in the salinity of freshwaters is known to cause adverse effects on the fitness and survival of many aquatic organisms, however the osmoregulatory mechanisms underlying these effects are still poorly understood. Moreover, it has been proved that different ions can have different toxicities to aquatic organisms, but most of the research has focused exclusively on Na^+ and Cl^-. Thus, more investigations on the potential effects of increasing concentrations of other specific ions (e.g. K^+, SO_4^{2-}) are needed. The impact of salinization on freshwater biodiversity can alter ecosystem functioning, although only a few functions have been studied (e.g. leaf litter decomposition). Also, freshwater salinization can affect the delivery of ecosystem services and have direct economic (e.g. infrastructure corrosion) and social (e.g. human health) impacts that have rarely been assessed. The management of this urgent environmental issue needs to be improved through mitigation (e.g. backfilling of mine tailings), remediation (e.g. enhanced wastewater treatment), prevention (e.g. using alternative deicers for roads) and monitoring (e.g. estimating salt loads to freshwater ecosystems) actions.

Key words: salinity, pollution, sub-lethal effects, resource extraction, agriculture, climate change, ecosystem functioning, wastewater treatment, human health

RESUMEN

Revisión de los avances recientes y los desafios futuros en la salinización de los ecosistemas de agua dulce

A pesar de ser un fenómeno mundial que puede tener importantes consecuencias ecológicas, económicas y sociales, la salinización de los ecosistemas acuáticos (es decir, el aumento de las concentraciones de iones en sus aguas) ha sido poco estudiada en comparación con otros problemas ambientales. Sin embargo, está recibiendo una atención cada vez mayor, con importantes avances científicos realizados durante las últimas dos décadas. Aquí reviso el conocimiento actual sobre el tema y propongo futuras direcciones de investigación en el contexto del bienestar humano y el cambio global. La salinización de los ecosistemas acuáticos es causada por una amplia gama de actividades humanas, siendo la agricultura y la extracción de recursos las más ampliamente documentadas. Diferentes estudios sugieren que podría estar afectando alrededor de 1/3 de los cuerpos de agua dulce, y este número probablemente aumentará en el futuro debido a los cambios en el clima y los usos del suelo. Se sabe que el aumento en la salinidad de las aguas dulces causa efectos adversos en la condición física y la supervivencia de muchos organismos acuáticos, sin embargo, los mecanismos osmorreguladores que subyacen a estos efectos aún no se conocen bien.

Además, se ha demostrado que diferentes iones pueden tener diferentes toxicidades para los organismos acuáticos, pero la mayoría de las investigaciones se han centrado exclusivamente en los iones Na^+ y Cl^-. Por lo tanto, se necesitan más investigaciones sobre los efectos potenciales de otros iones (por ejemplo, K^+, SO_4^{2-}). El impacto de la salinización en la biodiversidad de agua dulce puede alterar el funcionamiento de los ecosistemas acuáticos, aunque solo se han estudiado algunas funciones (por ejemplo, la descomposición de la hojarasca), y podría afectar a la provisión de servicios ecosistémicos. Finalmente, la salinización de agua dulce también puede tener impactos económicos directos (p. ej. corrosión de la infraestructura) y sociales (p. ej. problemas de salud humana) que rara vez se han evaluado. La gestión de este urgente problema ambiental debe de mejorarse a través de acciones de mitigación (p. ej. retorno de los residuos mineros a los huecos de las explotaciones mineras), remediación (p. ej. tratamiento mejorado de aguas residuales), prevención (p. ej. uso de descongeladores alternativos para carreteras) y monitoreo (p. ej. estimación de las aportaciones de sal a los ecosistemas de agua dulce).

Palabras clave: *salinidad, contaminación, efectos sub-letales, extracción de recursos, agricultura, cambio climático, funciones ecosistémicas, tratamiento de aguas residuales, salud humana*

INTRODUCTION

Freshwater salinization can be defined as an increase in the ion concentration of freshwater ecosystems. However, it should be noted that salinization not only changes the total concentration of total dissolved ions, but it usually changes ion ratios as well. Marine and freshwaters greatly differ in the concentration and proportion of ions, with the former being dominated by Na^+ and Cl^- and the latter by Ca^{2+} and HCO_3^- (Margalef, 1986). This is important because aquatic organisms have evolved under relatively constant ion concentrations and ratios. As Ranking & Davenport (1981) stated: "The body fluids of living organisms are dilute salt solutions reflecting the origin of life in the sea". Thus, it is not surprising that dissolved salts (i.e. ions) concentrations in the water are one of the main factors controlling life on Earth (Bradley, 2008) and probably in other planets (Marion *et al.*, 2003). Aquatic animals need to maintain a balance between the osmotic concentrations of their internal media (i.e. cells and body fluids) and the media in which they live (i.e. water) (Rankin & Davenport, 1981; Larsen *et al.*, 2014). Most aquatic organisms have the ability to control internal ion concentrations (i.e. osmoregulation) to some limit. Pass this limit osmoregulation breaks down, and they start to osmoconform (Williams & Feltmate, 1992). Eventually, cells are damaged and die, leading to stress and mortality of the individuals. Consequently, aquatic communities are strongly shaped by the salinity of their surrounding water. For example, the species richness of brackishwater environments is largely constrained by salinity fluctuations (Remane & Schlieper, 1971; Attrill & Rundle, 2002), which have important evolutionary implications for the physiology of salt and water balance (Larsen *et al.*, 2014). Also, background salinity (i.e. not modified by human action) is known to limit species richness in streams (Egglishaw & Mackay, 1967; Minshall & Kuehne, 1969). This is especially evident in arid streams, which can comprise endemic fauna with salinity adaptations (Sánchez-Fernández *et al.*, 2008; Millán *et al.*, 2011). For example, Arribas *et al.* (2013) found 4 cryptic species within the *Enochrus falcarius* species complex that could have emerged from allopatric speciation caused by the isolation of saline habitats (i.e. surrounded by a matrix of freshwater habitats).

Freshwater salinization is caused by a wide variety of activities (Fig. 1). The most documented are: agriculture and pasture (Allison *et al.*, 1990; Williams, 2001a; Halse *et al.*, 2003), resource extraction (Pond *et al.*, 2008; Cañedo-Argüelles *et al.*, 2012; Warner *et al.*, 2013) and the use of salts as deicing agents for roads (Kaushal *et al.*, 2005; Corsi *et al.*, 2010). Urban and industrial wastewaters can also contribute to increase ion concentrations in receiving water bodies, but they have been suggested to be less important (Meybeck, 2003; Berger *et al.*, 2017).

The increase of salts in freshwater ecosystems is known to greatly reduce aquatic biodiversity (Cañedo-Argüelles *et al.*, 2013), being among the top causes of biological degradation in rivers and streams (Vander Laan *et al.*, 2013; De Castro-Català *et al.*, 2015) and one of the stressors with

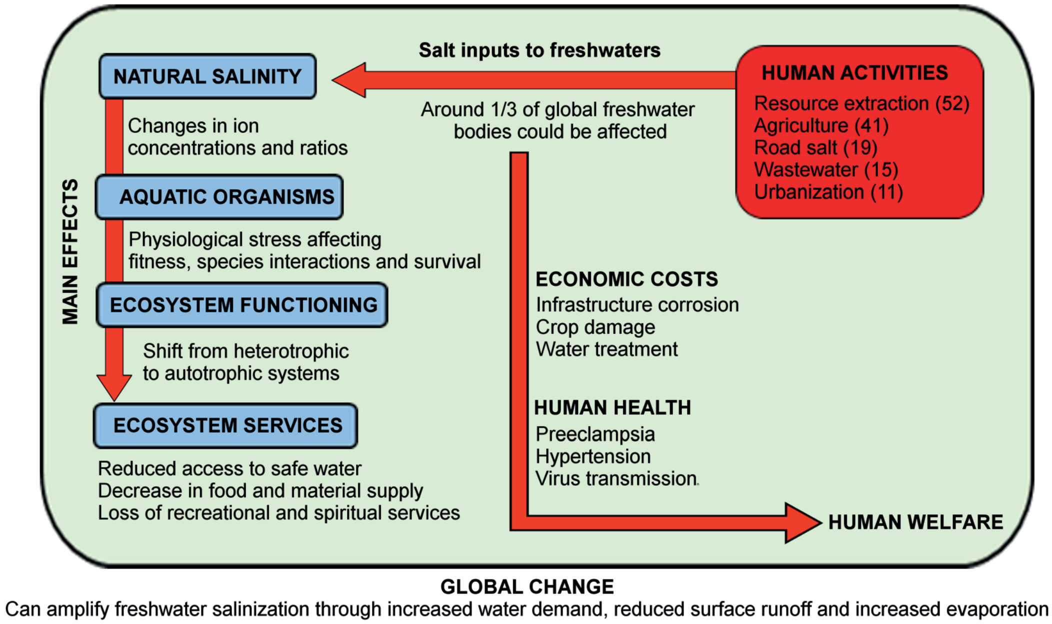

Figure 1. Schematic diagram of the main topics covered in this paper. Human activities modify the ion concentrations and ratios of freshwater ecosystems through point source and diffuse salt pollution (i.e. freshwater salinization). This has an impact on aquatic organisms through physiological stress that leads to changes in ecosystem functioning and the delivery of ecosystems services. Additionally, freshwater salinization have direct economic costs and effects on human health that, combined with the impacts on ecosystems, affect human welfare. The impacts of freshwater salinization on ecosystems and human welfare will very likely be amplified in the future due to global change, since the salt dilution capacity of freshwater bodies will be reduced. Next to each human activity, in brackets, I show the number of cited papers referred to each of them. *Diagrama esquemático de los principales temas tratados en este artículo. Las actividades humanas modifican las concentraciones y proporciones de iones de los ecosistemas de agua dulce (fenómeno denominado salinización de los ecosistemas acuáticos). Esto tiene un impacto en los organismos acuáticos a través del estrés fisiológico que conduce a cambios en el funcionamiento del ecosistema y la prestación de servicios ecosistémicos. Además, la salinización de los ecosistemas acuáticos tiene costos económicos y efectos directos en la salud humana que, combinados con los impactos en los ecosistemas, afectan el bienestar humano. Es muy probable que los impactos de la salinización en los ecosistemas y el bienestar humano se amplifiquen en el futuro debido al cambio global, ya que la capacidad de dilución de sal de los cuerpos de agua dulce se reducirá. Junto a cada actividad humana, entre paréntesis, muestro el número de documentos citados referidos a cada una de ellas.*

the strongest potential impact on aquatic organisms (Velasco *et al.*, 2019). Biodiversity reduction occurs at both taxonomic and functional levels (Piscart *et al.*, 2006; Szöcs *et al.*, 2014; Suárez *et al.*, 2017), and it decreases more rapidly after a certain threshold is reached. According to field studies, the salinity threshold at which most freshwater species are extirpated could be around 4–10 g/L (Williams *et al.*, 1990; Pinder *et al.*, 2005), but it can vary widely among taxa (Kefford *et al.*, 2012). Salt pollution had been already acknowledged as a water quality problem since the 1920s (Meybeck & Helmer, 1989). Also, Dr. Robert A. Berner showed in 1971 that roughly one-third of the sulphate carried by rivers was derived from the activities of man. However, freshwater salinization was largely neglected by most water resource managers, conservationists and limnologists for decades (Williams, 2001b). Several papers with a wide focus on the topic have been published during the last 18 years (e.g. Williams 2001a; Herbert *et al.*, 2015; Cañedo-Argüelles *et al.*, 2016; Dugan *et al.*, 2017; Kaushal *et al.*, 2018; Cañedo-Argüelles *et al.*, 2019), but it is still receiving much less attention than other environmental issues (Fig. 2). In my opinion, this could be related with the common belief among scientists that this is a local issue occurring almost exclusively in arid regions. Additionally, the fact that salts occur naturally in freshwaters has probably prevented scientist and managers from considering salts as pollutants (Gorostiza & Sauri, 2019). Here I review the latest advances in freshwater salinization research, paying special attention to river ecosystems. The aim of this paper is to promote future investigations on the topic by identifying current knowledge gaps and management needs.

THE SPATIAL EXTENT OF FRESHWATER SALINIZATION

How many freshwater ecosystems of the world are salinized? Are there freshwater salinization hotspots? Given the available information, it is very difficult to answer these questions precisely. The global spatial extent of freshwater salinization remains unknown but available data suggests that it is affecting a large proportion of freshwater ecosystems. For example, Kausahl *et al.* (2018) estimated that salinization had impacted 37 % of the drainage area of the contiguous USA, observing statistically increasing trends at 39 % of sites for specific conductance, 34 % of sites for sodium, 29 % for calcium, 33 % for magnesium, and 36 % for potassium concentrations. Similarly, Estévez *et al.* (2019) showed that around 27 % of the streams in Spain were salinized. Finally, Le *et al.* (2019) found statistically increasing trends in electrical conductivity (EC) for around 80 % of the streams in Germany.

Among the different causes of freshwater salinization, agriculture seems to be the most important (Estévez *et al.*, 2019). In 2001 Williams (2001a) reported that over 10 % of all irrigated land was already damaged and around 25 % of the world water bodies could be salinized by agriculture. Other estimates are considerably higher and indicate that around 20–50 % of all irrigated lands may be salt-affected (Pitman & Läuchli, 2002). Since agricultural land is rapidly expanding (Foley *et al.*, 2005), these numbers will likely increase in the future. Resource extraction is another major cause of freshwater salinization through both the disposal of produced waters and the weathering of exposed rocks. For example, shale gas extraction can produce waters up to 7 times more saline than seawater (Vengosh *et al.*, 2014). This salinization source will also become more important in the future (Ferrar *et al.*, 2013). In the USA shale gas extraction is expected to increase threefold and will account for nearly half of all natural gas produced by 2035 (Entrekin *et al.*, 2011). This might lead to huge volumes of salinized water entering freshwaters through shale gas wastewater disposal, spills and application to roads (Olmstead *et al.*, 2013; Patterson *et al.*, 2017; Tasker *et al.*, 2018). Rozell & Reaven (2012) projected that if only 10 % of the Marcellus shale region was developed, this would result in a volume of contaminated water equalling a few thousand Olympic-sized swimming pools. Potash mining is another major source of salts in rivers and streams. For example, due to the potash mines in Alsace, the estimated chloride load increased in the Rhine River from less than 50 kg/s to more than 300 kg/s in the 1960's (Meybeck & Helmer, 1989). In Germany, the River Werra registered peak Cl^- concentrations of 30 g/L when flowing near the salt mines

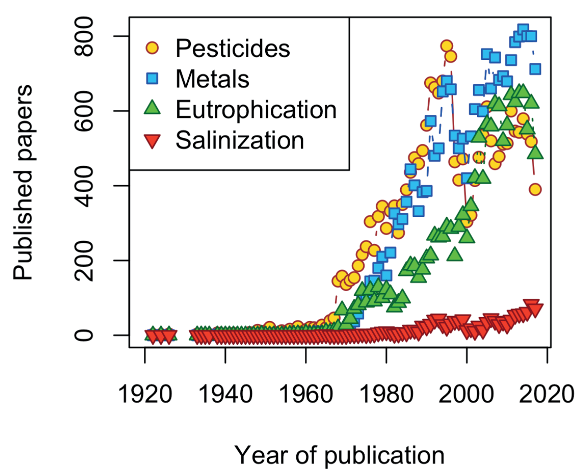

Figure 2. Number of papers published by year (not cumulative) on pesticide and metal pollution, eutrophication and salinization of freshwaters. The data was retrieved from the Web of Science website on the 26th of Februarys of 2018 after searching for the terms "freshwater pesticide", "freshwater metal", "freshwater eutrophication" and "freshwater salinization", respectively. *Número de artículos publicados por año (no acumulativos) sobre contaminación de plaguicidas y metales, eutrofización y salinización de ecosistemas acuáticos. Los datos se obtuvieron de la página web Web of Science el 26 de febrero de 2018 después de buscar los términos "freshwater pesticide", "freshwater metal", "freshwater eutrophication" and "freshwater salinization", respectivamente.*

(Coring & Bäthe, 2011). Road salt is emerging as another important source of salts (mainly NaCl) into freshwaters in the cold regions of the world (Lofgren, 2001; Ruth, 2003; Kaushal *et al.*, 2005) and it is tightly linked to urbanization (Novotny *et al.*, 2008; Kaushal *et al.*, 2018). Due to the application of NaCl as a deicing agent in roads Cl^- concentrations exceeded U.S. Environmental Protection Agency (USEPA) acute (860 mg/L) and chronic (230 mg/L) water-quality criteria at 55 and 100 % of monitored sites in 11 watersheds of Milwaukee (USA), respectively (Corsi *et al.*, 2010). Road salt application might also increase in the future due to urban expansion. In the US, it has increased from 0.20 to 24.5 million metric tons per year in seven decades (Coldsnow *et al.*, 2017a). Although there is not much information about it, industrial effluents are likely to further contribute to freshwater salinization (Berner, 1971; Piscart *et al.*, 2005). For example, in the US the chemical industry accounted for about 37 % of total salt sales in 2017 (Ober, 2018). Finally, chemical weathering of carbonate rich urban infrastructure can be a potential source of Ca^{2+}, Mg^{2+} and other major ions to streams (Kaushal *et al.*, 2014, 2015, 2017; Bird *et al.*, 2018).

Arid lands with naturally saline groundwater are a good example of freshwater salinization hotspots and show how this issue is probably occurring more widely than it is being reported. Taking Australia as an example (Allison *et al.*, 1990; Williams, 2001a), it has been proved that forest clearing for agriculture and pasture in these areas leads to freshwater salinization trough rising water tables (that are naturally saline) due to a reduction in the soil water retention capacity. However, naturally saline groundwaters are not restricted to Australia, they occur worldwide and are especially abundant in Asia (Van Weert *et al.*, 2009). For example, the largest area with high groundwater salinity is found in West and Central Asia (Van Weert *et al.*, 2009) and 23 % of the 30 000 km^3 of groundwater stored in the Indo-Gangetic Basin (IGB) alluvial aquifer has a salinity greater than 1 g/L (MacDonald *et al.*, 2016). Thus, it is reasonable to assume that, similarly to Australia, freshwater salinization is widely occurring in agricultural areas of Asia, although only a few cases have been reported (e.g. the Aral Sea region: Létolle & Chesterikoff, 1999; Micklin, 2007; Aladin *et al.*, 2009). Additionally, salinization of Asian rivers and streams have been associated with urban development (Bhatt & McDowell, 2007) and chemical, paper and textile industries (Lokhande *et al.*, 2011; Nirgude *et al.*, 2013). Examples of freshwater salinization can also be found in other regions of the world. In South America, very high salt loads into the Amazon River have been reported due to oil extraction (Moquet *et al.*, 2014). Also, increases in the salinity of freshwaters have been associated with salt factories (González Achem *et al.*, 2015) and drainage canals (Bazzuri *et al.*, 2018) in Argentina and salt extraction (Akomolafe & Onwusiri, 2017), agriculture (Scherman *et al.*, 2003) and industry (Dikio, 2010) in Africa.

Increased water demand (Vörösmarty *et al.*, 2000) will exacerbate freshwater salinization, since it leads to reduced salt dilution capacity of water bodies. For example, very high concentration of salts toxic to aquatic fauna have been found in saline lakes due to lowering water levels (Aladin *et al.*, 2009; Herbst *et al.*, 2013). Also, a combination of sea-level rise and seawater intrusion caused by groundwater over-exploitation for agriculture is leading to the salinization of coastal freshwater ecosystems (Mirza, 1998; Craft *et al.*, 2009; Mahmuduzzaman *et al.*, 2014; Herbert *et al.*, 2015; Pereira *et al.*, 2019). Blankespoor *et al.* (2012) predicted that freshwater wetlands would be lost at an average rate of 64 % to submergence and conversion to saline systems following a 1-m rise in sea level, with the highest regional loss rates in the Middle East and North Africa (100 %), Latin America and the Caribbean (74 %), Sub-Saharan Africa (72.5 %), and East Asia and the Pacific (62.2 %). Additionally, climate change-induced extreme precipitation (IPCC, 2013) might increase the frequency of river embankment overtopping, thereby flooding surrounding water bodies with saline river water (Karim & Mimura, 2008). Overall, it is clear that freshwater salinization will expand and increase in the future. Olson (2019) predicted that by 2100 12 % of the streams in the USA will have an EC greater than 2 mS/cm and the number of streams unusable for irrigation will double due to the combined effect of land use and climate change. Also, Le *et al.* (2019) predicted that EC will increase by 50 % in 5–10 % of the streams in Germany in the period from 2070 to 2100.

OSMOREGULATION AND ION TOXICITY

We know that different ions are regulated by different, but functionally similar, transporters (Griffith, 2017) and that body size and phylogeny are probably important factors determining salt sensitivity through their effect on the ion uptake rates of aquatic organisms (Kefford *et al.*, 2012; Poteat & Buchwalter, 2014; Scheibener *et al.*, 2016). However, there is still much uncertainty regarding basic osmoregulatory processes in freshwater fauna and how they relate to toxicity, especially for some ions (e.g. SO_4^{2-}) and taxa (e.g. aquatic insects, amphibians) (Hopkins & Brodie, 2015; Scheibener *et al.*, 2017). For example, it is not fully understood why some mayfly species experience significant mortalities at ECs lower than those of their haemolymph (Dowse *et al.*, 2017; Kefford *et al.*, 2019).

Although NaCl has received most of the scientific and public attention as a dominant form of salt pollution, other salts can greatly contribute to salinization (Kaushal *et al.*, 2018). Cormier *et al.* (2013) found that HCO_3^- and SO_2^{4-} concentrations were greater than Cl^- concentrations in most catchments of West Virginia (USA). Also, Moore *et al.* (2017) proved that concrete weathering can be an important source of Ca^{2+} and SO_2^{4-}. This is important because available studies show that not all ions are equally toxic to freshwater biodiversity. For example, Potapova and Charles (2003) found that some diatom species in USA streams were strongly affected by the proportions of individual cations and anions, and by the ratio of monovalent to divalent cations. Scheibener *et al.* (2016) found that the dominant anion could profoundly influence Na^+ uptake rate in the caddisfly *Hydropsyche betteni*. Kunz *et al.* (2013) reported different sensitivities of different aquatic invertebrates to reconstituted water with different ionic matrices. Clements and Kotalik (2016) found that $MgCl_2$ had toxic effects to stoneflies and mayflies below the EPA chronic chloride quality standard. Among the different ions, K^+ seems to be the most toxic to freshwater fauna (Mount *et al.*, 1997, 2016; Griffith, 2017) and SO_2^{4-} seems to be more toxic than Cl^- (Soucek & Dickinson, 2015; Clements & Kotalik, 2016). On the contrary, water hardness (i.e. dissolved $CaCO_3$ and $MgCO_3$) can ameliorate the toxic effects of other ions (Soucek & Kennedy, 2005; Soucek, 2007b; Van Dam *et al.*, 2010; Elphick *et al.*, 2011; Soucek *et al.*, 2011), except K^+ (Mount *et al.*, 2016; Soucek *et al.*, 2018). Also, K^+ could ameliorate SO_2^{4-} toxicity in cladocerans, midges, mussels and fish (Wang *et al.*, 2016). Overall, it is clear that the impact of salinization on freshwater ecosystems will need to be assessed within the context of multiple interacting ions (Bogart *et al.*, 2019; Schulz & Cañedo-Argüelles, 2019).

Ion toxicity could vary greatly among taxa. For example, Castillo *et al.* (2018) performed a meta-analysis of lethal salinity concentrations and found that microinvertebrates were more sensitive to salinity than vertebrates and macroinvertebrates. Hintz & Relyea (2017) showed that $MgCl_2$ did not affect rainbow trout growth significantly, whereas it had been shown to greatly affect amphibians (Hopkins *et al.*, 2013). Also, in a review of the German literature on freshwater salinization, Schulz & Cañedo-Argüelles (2019) reported the following salinity sensitivity of freshwater fauna (from more to less sensitive): fish > macroinvertebrates > phytoplankton > macrophytes > diatoms. Overall, available data suggest that amphibians, fish and certain macroinvertebrates (e.g. some caddisflies and stoneflies) are among the most sensitive to salinity (Cañedo-Argüelles *et al.*, 2013). However, the comparisons of salt toxicity across organisms are scarce. Moreover, it is still unclear whether the different response of organisms to salt toxicity results from total ionic concentration, the concentration of certain ions or the relative ratio of ions to each other (van Dam *et al.*, 2014; Scheibener *et al.*, 2017).

INTERACTION WITH OTHER STRESSORS

Freshwater ecosystems are affected by different point-source and diffuse pollution sources as well as hydrological and physical alterations, thus being subjected to multiple and interacting stressors (Meybeck, 2003; Walsh *et al.*, 2005; Ormerod *et al.*, 2010; Jackson *et al.*, 2016). Given that salinization is caused by a wide variety of human activities, its potential interacting stressors are many and can interact in multiple ways (Velasco *et al.*, 2019). Here I focus on those that have been most widely reported in the literature.

Alkalization

Increasing concentrations of dissolved salts with strong bases and carbonates, together with the displacement of base cations on soil exchange sites, can increase the pH of fresh water over time, linking salinization to alkalization (Kaushal *et al.*, 2018). This is mainly associated with the weathering of carbonate lithology by acid rain, which is leading to increasing trends in alkalization (Kaushal *et al.*, 2013). Alkalization can have a wide variety of effects on freshwater ecosystems, e.g. increasing dissolved organic carbon (Steele & Aitkenhead-Peterson, 2013), contributing to ammonia toxicity (Erickson, 1985; Boyd *et al.*, 2016) and stimulating nitrate production by microbial nitrification (Kaushal *et al.*, 2014). Although alkalinity alters the toxicity of different ions (Mount *et al.*, 2016), there is barely any information on the potential synergistic, antagonistic and additive effects of alkalization and salinization. Given that both stressors generally co-occur, this is an issue that deserves to be studied. Moreover, the interaction between alkalization and salinization could promote ocean acidification (Kaushal *et al.*, 2018).

Metals

Salinity has been widely reported to ameliorate the toxicity of most metals (Cañedo-Argüelles *et al.*, 2013). This is probably related with lower salinities leading to a greater bioavailability of toxic metal forms (Hall & Anderson, 1995) and to a modification of osmoregulation leading to higher metal uptake (Grosell *et al.*, 2007). Available studies suggest that different ions interact in different ways with metals (Boyd *et al.*, 2016), affecting their toxicity (e.g. Ca^{2+} can be more effective than Mg^{2+} in ameliorating Cu^{2+} toxicity). However the mechanisms behind the interactions of metals with specific ions are still poorly understood and they can vary across taxa. For example, Scheibener *et al.* (2016) found that silver (Ag) and copper (Cu), known to be antagonistic to Na^+ uptake in other aquatic taxa, did not exhibit this effect in different caddisfly and mayfly species. Also, Poteat and Buchwalter (2014) showed that Ca^{2+} uptake did not appear to be compromised by Cd or Zn exposure in caddisfly and mayfly species, whereas shared uptake pathways between Ca^{2+} and the heavy metals Cd and Zn had been reported for fish, crustaceans and molluscs. Although salts can ameliorate metal toxicity, salinization can also promote metal pollution by displacing metals from ion exchange sites on soils and sediments and by increasing their bioavailability (Kaushal *et al.*, 2013; Schuler and Relyea, 2018; Kaushal *et al.*, 2019). Overall, more information is needed on the effect of salts on the accumulation of metals in freshwaters and their potential impact on ecosystem functioning and service provision (Schuler & Relyea, 2018).

Pesticides

Given that pesticides often co-occur with increased ions concentrations in agricultural regions (Williams, 1987), pesticides and salinity can potentially interact to impact aquatic fauna. However, the few available studies show contradicting results. Song *et al.* (1997) exposed salt marsh mosquitoes to four insecticides (aldicarb, dimethoate, imidacloprid, and tebufenozide) and found less mortality under isosmotic conditions than under hyperosmotic conditions (probably due to the energetic costs associated with osmoregulation). Also, the toxicity of organophosphate pesticides has been reported to increase with increasing salinity (Hall & Anderson, 1995). On the contrary, other studies found no interaction between both stressors (Schäfer *et al.*, 2011, 2012; Szöcs *et al.*, 2012), suggesting that their combined effects might be exclusively additive.

Eutrophication

Salts can mobilize organic nitrogen, ammonium, and phosphorus via ion exchange (e.g. by the reduction of NH_4^+ by Na^+) and stimulate eutrophication (Duan *et al.*, 2012; Duan & Kaushal, 2015; Kaushal *et al.*, 2013, 2017; Haq *et al.*, 2018). This can promote the proliferation of cyanobacteria blooms, not only because the direct effects of salts on algae (Cañedo-Argüelles *et al.*, 2017), but also because the reduction of salt sensitive cladocerans which feed on algae (Jeppesen *et al.*, 2007, 2015; Brucet *et al.*, 2010). Since salinity and nutrient pollution often originate from the same sources (Walsh *et al.*, 2005), the contribution of salinity to eutrophication (e.g. through algal proliferation) deserves further attention.

Drought

Several studies suggest that salinity tolerance evolved during periods of aridification, probably as a by-product of adaptation to desiccation (Céspedes *et al.*, 2013; Arribas *et al.*, 2014; Pallarés *et al.*, 2016; Villastrigo *et al.*, 2018). Thus, the response of aquatic organisms to both stressors should be tightly linked. In a study of cross-tolerance to drought and salinity by aquatic beetles, Pallarés *et al.* (2017) found that salinity exposure enhanced desiccation resistance by activating water uptake mechanisms, whereas desiccation exposure produced water loss and haemolymph osmolality that decreased salinity tolerance. Concordantly, Suárez *et al.* (2017) found an antagonistic interaction between both stressors (i.e. the net effect of the two stressors was not as strong as the sum of their independent effects) on aquatic macroinvertebrates, probably because they were affecting the same set of species. However, synergistic interactions between drought and salinization could also be expected for salt sensitive species like certain mayflies, since reduced water flows and levels (i.e. in lotic and lentic ecosystems, respectively) might cause an increase in water temperature, thereby enhancing salt toxicity (Jackson & Funk, 2019). Also, the negative effects of water stress on fungal biomass can be amplified by salt pollution (Gonçalves *et al.*, 2019a). Due to a combination of water withdrawals for human consumption (Foley *et al.*, 2005) and climate change (Gudmundsson *et al.*, 2018), both stressors will very likely intensify in the future in arid regions of the world (Jones *et al.*, 2018). This could have important consequences for the aquatic ecosystems, since they could be colonized and dominated by a few salinity and/or drought tolerant species (Kefford *et al.*, 2016) and ecosystems functions such as organic matter decomposition could be modified (Gonçalves *et al.*, 2019a).

SUB-LETHAL EFFECTS AND TROPHIC INTERACTIONS

Salinization causes a decline in freshwater biodiversity, with salinity tolerances varying across ions, species and regions (Kefford *et al.*, 2012; Cañedo-Argüelles *et al.*, 2013; Herbert, 2015; Castillo *et al.*, 2018). However, beyond causing mortality, increased salt concentrations can have sub-lethal effects on freshwater organisms that have been less studied and that might modify ecosystem functioning through alterations in individual fitness and trophic interactions. Johnson *et al.* (2015), Herbst *et al.* (2013) and Hintz & Relyea (2017) found that salinization reduced growth of the mayfly *Neocloeon triangulifer*, the damselfly *Enallagma clausum* and the rainbow trout, respectively. This was very likely related with an energetic demand imposed by ion transportation for maintaining homeostasis (Romano *et al.*, 2017; Buchwalter *et al.*, 2019). Also salinization can trigger behavioural responses such as increased drift of stream insects to reach more suitable habitats (Clements & Kotalik, 2016; Beermann *et al.*, 2018).

There are several potential impacts of sub-lethal responses to salinization on trophic interactions. Increased salinity can trigger trophic cascades by promoting algal growth through a reduction in the feeding efficiency of herbivorous invertebrates (Soucek, 2007a; c), thereby promoting eutrophication (Jeppesen *et al.*, 2007; Hintz *et al.*, 2017; Schuler *et al.*, 2017; Schulz & Cañedo-Argüelles, 2019). Additionally, freshwater salinization can affect species co-existence. For example, Stănescu *et al.* (2017) showed that increased salinity could benefit the toad *Pelobates syriacus* over the sympatric and endangered *P. fuscus*, and Bray *et al.* (2019) found that the interaction between salt tolerant and sensitive communities could significantly affect the response of some invertebrate species to increased salinity in a mesocosm experiment. Concordantly, Arribas *et al.* (2019) argued that the dominance of certain species in saline habitats (i.e. saline species) could be more related with avoiding competition or predation than with physiological adaptations. The modification of species interactions by salinization should be also analysed from a meta-community perspective, since salinized freshwater bodies could be poor in species due to a limitation in the sources of salt tolerant organisms (i.e. naturally saline sites) nearby (Gutiérrez-Cánovas, *et al.*, 2019). Finally, parasitism is another interaction that deserves to be studied within the frame of freshwater salinization. Buss and Hua (2018) found that NaCl increased tadpole susceptibility to parasites (i.e. trematodes), although this was dependent on the presence of tadpole predators (i.e. damselflies). Overall, salinized freshwater ecosystems could experience a reduction in food chain length due to constrained diversity of consumers and trophic resources in local food webs (East *et al.*, 2017).

The response of organisms to salinization might depend on the background salinity at which their populations have evolved (Kefford *et al.*, 2016). For example, the microcrustacean *Daphnia pulex* is known to rapidly evolve salt tolerance (Coldsnow *et al.*, 2017a), which is transferred to sub-sequent generations through methylation (Jeremias *et al.*, 2018). Also, intra-specific differences in salt sensitivity were found by Sala *et al.* (2016), who showed that individuals of the net-spinning caddisfly *Hydropsyche exocelleta* from low salinity streams built a lower number and less symmetric nets than a population coming from a salt-polluted stream. Salinity adaptation could have associated trade-offs modifying the organism performance. Coldsnow *et al.* (2017b) showed that increased salt exposure ablated the circadian clock of *D. pulex*, potentially affecting key behaviours like diel vertical migration. Adaptations to salinization could also play an important role in maintaining ecosystem stability, e.g. by mitigating trophic cascades (Hintz *et al.*, 2019). However, salinity adaptations need further exploration, since some studies have found no evidence of enhanced salinity resistance after multigenerational exposure to increased salt concentrations (Loureiro *et al.*, 2015; Venancio *et al.*, 2019).

EFFECTS ON ECOSYSTEM FUNCTIONING AND HUMAN WELFARE

Freshwater salinization can have important impacts on ecosystem functioning. For example, in wetlands salinization can alter biochemical cycles, leading to eutrophication and reduced carbon storage (Herbert *et al.*, 2015). In lakes, it can enhance water stratification, promoting phosphorous release from the sediment through the creation of anoxic conditions (Koretsky *et al.*, 2012). In rivers and streams changes in water salinity can significantly alter riparian-stream connections through different pathways (e.g. soil and stream decomposers and plant salt uptake) (Entrekin *et al.*, 2019). For example, salinization has been shown to diminish organic matter inputs and processing due to the reduction of riparian vegetation cover (Ladrera *et al.*, 2017) and leaf litter breakdown by invertebrates (Schäfer *et al.*, 2012; Cañedo-Argüelles *et al.*, 2014), respectively. The effect of salinity on microbial decomposition is less clear. On one hand, some studies have reported that microorganism-mediated leaf litter breakdown could be most efficient at intermediate salinities (Cañedo-Argüelles *et al.*, 2014; Sauer *et al.*, 2016). On the other hand, Canhoto *et al.* (2017) showed that trade-offs between growth and sporulation can maintain fungal growth and decomposition at high levels along a wide NaCl gradient (0–16 g/L) and Gonçalves *et al.* (2019b) found no effect of salinization on fungal sporulation in strains coming from sites with different background salinities (0.01 vs. 6 g/L). Overall, based on information coming from arid streams (Millán *et al.*, 2011; Berger *et al.*, 2019), salinized streams are likely to act as net carbon exporters due to a decrease in allochthonous organic matter inputs and decomposition and an increase in algal production and eutrophication.

All the impacts of salinization on biodiversity and ecosystem functioning might affect the services that freshwater ecosystems provide (Berger *et al.*, 2019). For example, the combined effects of salinization and drought led to the

Figure 3. Impacted stream (Torrent de Soldevilla, Catalonia, Spain) by potash mining (picture by Ruben Ladrera, Universidad de La Rioja). Riparian vegetation is absent from the margins of the stream. The mine tailing (mainly composed by NaCl) can be seen in the back of the picture. *Arroyo impactado (Torrent de Soldevilla, Cataluña, España) por la minería de potasa (foto de Rubén Ladrera, Universidad de La Rioja). La vegetación ribereña está ausente de los márgenes del arroyo. El relave de la mina (compuesto principalmente por NaCl) se puede ver en la parte posterior de la imagen.*

collapse of fish populations in the Aral Sea, which were the main source of food and jobs for local residents (Micklin, 2007). Also, recreation and aesthetic values of freshwater ecosystems can be seriously compromised by salinization, due to a decline in habitat quality and biodiversity (Fig. 3). Finally, linked with ecosystem services, there are important economic and social costs associated with salt pollution of freshwaters (Cañedo-Argüelles *et al.*, 2016; Schuler *et al.*, 2019; Schulz & Cañedo-Argüelles, 2019). Saline waters corrode infrastructure (Wilson, 2004; Moore *et al.*, 2017) and increase water treatment costs (Honey-Roses & Schneider, 2012). Moreover, when salinity increases the water can become useless for agriculture, domestic consumption and many industrial uses (Williams, 2001b; Kaushal, 2016, Schuler *et al.*, 2019). Salinized water not only can become unpalatable and damage crops, it can also pose important risks to human health. In Bangladesh seawater intrusion in the coast is leading to health problems (e.g. preeclampsia, hypertension) and forced migrations of millions of people (Karim & Mimura, 2008; Khan *et al.*, 2011, 2014; Vineis *et al.*, 2011; Mahmuduzzaman *et al.*, 2014; Dasgupta *et al.*, 2015). Also, leaching metals from water infrastructure and sediments has contributed to increased lead concentrations in drinking water in the USA (Kaushal, 2016). In Spain, rivers affected by potash mining showed high concentrations of bromomethanes (Gorostiza & Sauri, 2017), which have been related with cancer in humans (Min & Min, 2016). Finally, the application of salts in roads to prevent icing can pose a risk to people with high blood pressure (Cooper *et al.*, 2014). This is important because raised blood pressure is a major risk factor for coronary heart disease and the leading risk factor for stroke (Ezzati *et al.*, 2002; He & MacGregor, 2007; Scheelbeek *et al.*, 2016). Besides risks associated with drinking water quality, freshwater salinization can affect human health through wind-borne dust (i.e. respiratory problems), virus transmission (e.g. promoting mosquito populations) and mental illness induced by environmental degradation (Jardine *et al.*, 2007).

MANAGEMENT STRATEGIES

Water managers and policy-makers have rarely been urged by society to manage freshwater salinization (Cañedo-Argüelles *et al.*, 2016), although there have been notable exceptions (Gorostiza, 2014; Gorostiza & Sauri, 2017). Thus, it is not surprising that cost-effective freshwater salinization prevention, remediation and mitigation strategies are generally lacking. The priority option to combat freshwater salinization should be to reduce salt loads at their origin. In the case of mining, wastes should be used to refill the excavated areas (i.e. backfilling) (Rauche *et al.*, 2001; Benzaazoua *et al.*, 2008) and vegetation should be planted to stabilize the soil and recover the natural landscape (i.e. phytostabilization) (Mendez & Maier, 2008; Fellet *et al.*, 2011). Brine collectors aimed at disposing mining waste waters and runoff into the sea (Martín-Alonso, 1994) do not seem to be a suitable long-term management option, since they can lead to saline water leaks (Gorostiza, 2014) and they can affect coastal ecosystems (Schulz & Cañedo-Argüelles, 2019). In the case of shale gas extraction, produced waters can be injected deep underground. However, most operators prefer to recycle and then send waste waters to approved industrial wastewater treatment facilities because it is less costly (Rozell & Reaven, 2012). Moreover, in some regions the availability of disposal wells might be limited (Ferrar *et al.*, 2013; Vidic *et al.*, 2013). In the case of regions with saline groundwaters, the salinization caused by land clearing can be ameliorated by lowering groundwater tables through water pumping (Williams, 2001b) and the plantation of salt tolerant trees in areas where groundwater is accessible to their roots (Queensland Government, 2011). Agricultural practices should also be improved to use the amount of water that the crops require, preventing salts from accumulating in the soils and groundwater tables to rise (Gardner & Young, 1988; Pereira *et al.*, 2007; URS, 2014). Finally, salt applications in roads should be improved by selecting salts according to temperature/melting point and improving calibration/salt delivery techniques and the design of stormwater retention ponds (Kaushal, 2016; Snodgrass *et al.*, 2017). Additionally, alternative salts should be tested, although they tend to be more costly than NaCl (Jackson & Jobbágy, 2005). Finally, deicing technologies that ensure safe driving conditions while minimizing road salt application should be developed and implemented (Schuler *et al.*, 2019).

Another option for managing freshwater salinization is to improve wastewater treatment. Overall, wastewater treatment plants are not equipped to reduce dissolved ions (Entrekin *et al.*, 2011; Ferrar *et al.*, 2013), although multiple technologies (e.g. chemical precipitation, membrane technology, biological treatment) are available to do it (Shaffer *et al.*, 2013; Pinto *et al.*, 2016; Gibb *et al.*, 2017; Runtti *et al.*, 2018). This is mainly related with the high economic costs for the implementation and maintenance of these technologies (Karagiannis & Soldatos, 2008; Honey-Roses & Schneider, 2012). However, some promising advances are being made to reduce costs by using renewable energies (Mohamed & Papadakis, 2004; Nafey *et al.*, 2010) or biological treatments (Abou-Elela *et al.*, 2010; Smyntek *et al.*, 2017). Further technological developments coupled with rigorous cost-benefit analyses (including economic, environmental and social costs) are needed before remediation strategies can be widely implemented (Cañedo-Argüelles *et al.*, 2016; Wang *et al.*, 2018; Schuler *et al.*, 2019). Additionally, salt loads to freshwater ecosystems should be estimated to guide pollution mitigation and restoration efforts (Zuidema *et al.*, 2018; Schuler *et al.*, 2019). Complementarily, hyperspectral remote sensing could be used to identify heavily salinized areas (Pfitzner *et al.*, 2018). Finally, environmental flows should be established so the salt dilution capacity of rivers can be secured. For example, in the Murray Darling catchment (Australia) caps were placed on water diversions for consumptive uses and resulted in a recovery of aquatic biodiversity due to a reduction in salt concentrations (Paul *et al.*, 2018).

Finally, developing specific freshwater salinization monitoring programs is urgently needed (Schuler *et al.*, 2019). The concentration of dissolved ions in freshwaters can experience great intra and inter-annual variations due to changes in surface runoff (i.e. affecting the dilution capacity of continental waters and the diffuse salt inputs), evaporation and point-source discharges. Thus, it is very important to properly establish the monitoring periodicity of dissolved ions according to these fluctuations (Cañedo-Argüelles *et al.*, 2012; Timpano *et al.*, 2018). Since salinization can have multiple causes, stakeholders often deny responsibility (Gorostiza & Sauri, 2019). Thus, monitoring programs should include chemical fingerprinting (e.g. stable isotopes) to assess the contribution of the different human activities along the catchment to freshwater salinization (Otero & Soler, 2002; Osman *et al.*, 2017). Also, current biological indices do not seem to adequately respond to salt pollution (Cañedo-Argüelles *et al.*, 2012, 2017), thereby specific indices need to be developed (Schäfer *et al.*, 2011; Vander Laan *et al.*, 2013; Schulz, 2016; Halle *et al.*, 2017; Timpano *et al.*, 2018; Schulz & Cañedo-Argüelles, 2019). These indices should rely in the definition of reference conditions according to natural ion concentrations, which can vary widely depending on the catchment geology (Hawkins *et al.*, 2010; Griffith, 2014). Improved monitoring should be complemented with legally enforced standards for salinity and specific ion concentrations (both independently and in combinations) based on biodiversity and ecosystem protection (Cañedo-Argüelles *et al.*, 2016; Wang *et al.*, 2016; Cochero *et al.*, 2017; Bogart *et al.*, 2019; Schuler *et al.*, 2019).

OTHER FUTURE RESEARCH NEEDS

There are other topics that have been poorly investigated and deserve further attention. For example, there are potential sources of freshwater salinization that remain uninvestigated, such as abandoned metal mines (Mighanetara *et al.*, 2009), wildfires (Mast, 2013) and desalination brines (Kupsco *et al.*, 2017). Additionally, the dilution of naturally saline streams by freshwaters (e.g. coming from agricultural runoff) should be considered, since it threats the unique biodiversity that these ecosystems harbour (Velasco *et al.*, 2006; Sánchez-Fernández *et al.*, 2008; Millán *et al.*, 2011; Gutiérrez-Cánovas *et al.*, 2019). Overall, research should not only focus on the effects of the modification on the concentration of certain ions, but also on the modification of ion ratios. Also, future studies should assess the rate and duration of salt pollution because they can determine the response of freshwater organisms through resistance and resilience mechanisms (Hogan *et al.*, 2012; Cañedo-Argüelles *et al.*, 2014; Cooper *et al.*, 2014). Finally, freshwater salinization will very likely exacerbate in the future within the context of global change (Le *et al.*, 2019; Olson 2019). Thus, we need robust predictions of how the concentration of dissolved ions in freshwaters will change around the world as a result of increased agriculture (Foley *et al.*, 2005) and resource extraction (Krausmann *et al.*, 2009; Entrekin *et al.*, 2011), sea-level rise (IPCC, 2013; Mahmuduzzaman *et al.*, 2014) and reduced dilution capacity associated with lower river discharges (Dai *et al.*, 2009; Döll & Schmied, 2012) and lake levels (Jeppesen *et al.*, 2015). These predictions should be coupled with other global scale phenomena that can be influenced by freshwater salinization, such as ocean acidification (Doney, 2010; Gypens *et al.*, 2011; Kaushal *et al.*, 2018).

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