



**CURRENT EVIDENCE****Metals in coastal groundwater systems under anthropogenic pressure: a synthesis of behavior, drivers, and emerging threats**Tristan McKenzie <sup>1,\*</sup> Amy Moody,<sup>2</sup> João Barreira,<sup>3</sup> Xiaoyi Guo,<sup>4,5,6</sup> Anael Cohen,<sup>7</sup> Stephanie J. Wilson <sup>8</sup> Murugan Ramasamy<sup>9</sup>

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**Scientific Significance Statement**

Growing anthropogenic pressures along coastlines from population growth, urbanization, and climate change all contribute to increasing metal pollution risk in the coastal ocean. Coastal aquifers are a substantial reservoir of metals, yet due to their complex behavior and chemistry, metals are comparatively understudied in submarine groundwater discharge (SGD). This review synthesizes metal behavior, drivers, and processes in coastal groundwater systems in the context of climate change and human sources. In addition, we present results from a global coastal ocean metal contamination risk model, which demonstrates that the highest risk areas are tropical regions with high population density, SGD, and acid sulfate soils to drive and focus future research efforts.

**Abstract**

Submarine groundwater discharge (SGD) dynamically links land- and ocean-derived chemical constituents, such as metals, in the coastal ocean. While many metals are sediment-bound, changing environmental conditions, particularly along the coast, may lead to increased release of metals to their dissolved and more bioavailable form. Here, we review metal behavior, speciation, and drivers of mobilization in the coastal environment under anthropogenic influence. We also model global metal contamination risk to the coastal ocean via SGD considering anthropogenic and hydrogeologic pressures, where tropical regions with high population density, SGD, and acid sulfate soils (4% of the global coast) present the highest risk. Although most SGD studies focus

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on other analytes, such as nutrients, this review demonstrates the importance of considering SGD as a critical pathway for metals to reach the coastal ocean under rapidly changing environmental conditions.

Rapidly increasing anthropogenic pressure is driving declining coastal water quality and ecological health (Duarte 2014). Globally, population growth and urbanization rates are highest in coastal zones, where the human population is expected to exceed 1 billion by 2060 (Neumann et al. 2015). Decades of excess discharge from industrial, urban, and agricultural wastewater have resulted in substantial and lasting negative impacts to coasts (e.g., Rabalais et al. 2009).

Groundwater discharge is a critical influence on coastal chemistry across the land-ocean continuum. At the coastline, fresh groundwater meets with recirculated seawater in the coastal aquifer, or subterranean estuary (STE; Moore 1999), a highly biogeochemically active zone, where nutrients, organic matter, and metals can be transported to the coastal ocean. Submarine groundwater discharge (SGD) includes fresh meteoric water discharging to the coast and seawater recirculated through coastal sediments (Burnett et al. 2003). SGD has been widely recognized as a source of excess nutrients contributing to coastal eutrophication and poor water quality (e.g., Moore 1999). SGD is also an important source of metals (e.g., Beck et al. 2007; Kim and Kim 2011; Rodellas et al. 2014).

In coastal groundwater systems, metals have a wide range of sources, influencing processes, and applications. Metals can be grouped based on shared physical and chemical properties (Table 1), and include the alkali, alkali earth, transition, post-transition, metalloid, and rare-earth elements (REEs). For instance, many of the alkali, alkali earth metals, and REEs can be used to understand aquifer sources (e.g., Li, Sr, Ra, La, Tb), mixing of water masses (e.g., Na, K, Mg, Ca, Sr, Ce, Nd), and trace SGD (Ba, Ra, Sr, Si) (e.g., van de Flierdt et al. 2016; Garcia-Orellana et al. 2021; Mayfield et al. 2021). In comparison, many transition (e.g., Cd, Hg), post-transition metals and metalloids (e.g., Pb, As), and REEs (e.g., Gd, Ce) can be indicative of contamination from sewage, industrial, and agricultural waste (e.g., Beck et al. 2009; Trezzi et al. 2016; Johannesson et al. 2017; Prakash et al. 2021). Metals can act as important micronutrients for primary production (Tamborski et al. 2020a), enzymatic cofactors (Wang et al. 2016; Li et al. 2018), and tracers of circulation or transport (Bates et al. 2017), but they can also be toxic intrinsically or when concentrations exceed a threshold (e.g., Jaishankar et al. 2014). Both bioavailability and speciation, which influence toxicity, are driven by physicochemical and biological factors, including pH, redox conditions, and ligand availability (Worms et al. 2006; Bayen 2012). Furthermore, metals in solution have higher mobilities, and are often more bioavailable to primary and secondary producers compared to their particulate-bound forms (Miranda et al. 2022). Metal exposure

can disrupt biological functions such as reproduction and cognitive processes in both marine life and humans (Jaishankar et al. 2014).

Metal contamination is an escalating threat to coastal environments due to (1) changing aquifer conditions driven by climate change and (2) increasing coastal populations and urbanization. Here, we review metals transported by SGD in the context of anthropogenic change. Metals have been understudied in SGD compared to other analytes, such as nutrients, despite the important role they play in both natural biogeochemical cycling, as well as pollution. We also construct a risk assessment model to identify regions with the highest risk for coastal metal pollution via SGD in consideration of these pressures.

### *Metal behavior under rapidly changing conditions*

#### **Climate change–driven shifts to the coastal environment and STE**

Climate change impacts such as sea-level rise, warming temperatures, and precipitation changes are projected to disproportionately affect coastal areas (Neumann et al. 2015; Calvin et al. 2023), influencing key conditions for metal speciation in the STE. Coastal sediments and mangroves are frequently considered a metal sink, holding high concentrations of immobile sediment-bound metals (de Lacerda et al. 2022; Tang et al. 2022). Changes in salinity, pH, temperature, hydraulic gradients, land use, and redox conditions alter coastal metal speciation and adsorption behavior (e.g., Moore 1999), where SGD may become an increasingly significant source or sink of dissolved metals to the nearshore environment (Fig. 1).

#### *Sea-level rise and seawater intrusion*

Disruption of salinity gradients from sea-level rise and seawater intrusion will likely result in enhanced metal release to the coastal ocean (Moore 1999). While > 90% of total metal concentrations are sediment-bound, increasing ionic strength (salinity) can lead to the release of metals (e.g., Al, Cr, Mn, Cu, Co, Cd, V, Sr, Mo, Pb, Ra, Th, Ba, U, REEs) to solution (Beck et al. 2007, 2010a; Chen and Jiao 2008; Du Laing et al. 2008; Acosta et al. 2011; Liu et al. 2019b; Paffrath et al. 2020; Adyasari et al. 2021; Jia et al. 2021; Miranda et al. 2022). This is particularly true for cases where seawater intrusion pushes the STE further inland, exposing previously unexposed sediments to new, more saline, conditions, and releasing previously immobilized metals through processes such as sulfide desorption (Du Laing et al. 2008; Qu et al. 2020; Jia et al. 2021; Richardson et al. 2024). For instance, areas with reclaimed land are frequently a hotspot

**Table 1.** Main sources of metals to the coastal ocean and example applications in SGD work.

Metal group	Metal	Main coastal source(s)—natural	Main coastal source(s)—anthropogenic	Applications in SGD work	References
Alkali	Lithium (Li)	Aquifer lithology (i–ii)	Mining (iii); electronics (iii); aluminum production (iii); pharmaceuticals (iii)	Aquifer source (i); groundwater evolution (i), chemical weathering (i–ii); seawater–freshwater interactions (iv)	(i) Martin et al. (2020); (ii) Bagard et al. (2015); (iii) Barbosa et al. (2023); (iv) Russak et al. (2016)
	Sodium (Na)	Seawater (i–ii)	Mining (iii); wastewater (iii); fertilizers (iii); de-icing (iii)	Seawater–freshwater interactions (i–ii); anthropogenic inputs (iii)	(i) Nouayti et al. (2022); (ii) Salem et al. (2016); (iii) Kaushal et al. (2023)
	Potassium (K)	Seawater (i)	Mining (ii–iii); wastewater (ii); fertilizers (ii–iii); de-icing (ii)	Seawater–freshwater interactions (i, iii); anthropogenic inputs (ii–iii)	(i) Schiavo et al. (2006); (ii) Kaushal et al. (2023); (iii) Prakash et al. (2020)
	Cesium (Cs)	—	Nuclear fission product (i–iv)	Anthropogenic inputs (i–iv)	(i) Buesseler et al. (2018); (ii) Sanial et al. (2017); (iii) Takata et al. (2020); (iv) Kambayashi et al. (2021)
Alkali earth	Magnesium (Mg)	Seawater (i–ii)	Mining (iii); wastewater (iii); fertilizers (iii); de-icing (iii)	Seawater–freshwater interactions (i–ii); anthropogenic inputs (iii)	(i) Mayfield et al. (2021); (ii) Tillman et al. (2014); (iii) Kaushal et al. (2023)
	Calcium (Ca)	Seawater (i–ii); weathering (i–ii)	Mining (iii); wastewater (iii); fertilizers (iii); de-icing (iii); batteries (iv); paper production (iv)	Seawater–freshwater interactions (i–ii), anthropogenic inputs (iii–iv)	(i) Mayfield et al. (2021); (ii) Tillman et al. (2014); (iii) Kaushal et al. (2023); (iv) Prakash et al. (2020)
	Strontium (Sr)	Aquifer lithology (i); riverine sediments (ii)	Industry (i); metal processing (i); electronics (i); nuclear fission product (i)	Seawater–freshwater interactions (ii–iv); redox conditions (i); groundwater tracer (v–vi); anthropogenic inputs (i)	(i) Prakash et al. (2021); (ii) Rahaman and Singh (2012); (iii) Jørgensen et al. 2008; (iv) Musgrove and Banner (1993); (v) Beck et al. (2013); (vi) Chakrabarti et al. (2018)
	Barium (Ba)	Aquifer lithology (i–ii); riverine sediments (iii)	Industry (iii); agriculture (iii)	Groundwater tracer (iv–v); adsorption/release on Fe/Mn oxides (vi); desorption via ion	(i) Santos et al. (2011); (ii) Mayfield et al. (2021); (iii) Prakash et al. (2021); (iv) Moore

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**Table 1.** Continued

Metal group	Metal	Main coastal source(s)—natural	Main coastal source(s)—anthropogenic	Applications in SGD work	References
				exchange reactions (vi)	(1997); (v) Moore and Shaw (2008); (vi) Charette and Sholkovitz (2006)
	Radium (Ra)	Aquifer lithology (i–iii); riverine sediments (i–iii)	—	Groundwater tracer (i–iii); water residence times (iii); adsorption/release on Fe/Mn oxides (iv); desorption via ion exchange reactions (iv)	(i) Garcia-Orellana et al. (2021); (ii) Rama and Moore (1996); (iii) Moore (2000); (iv) Charette and Sholkovitz (2006)
Transition	Vanadium (V)	Aquifer lithology (i)	Mining (ii); fossil fuel production (ii); metal alloys (iii); wastewater (iv); agriculture (v)	Redox conditions (i, v–vi); anthropogenic inputs (ii–v)	(i) O’Connor et al. (2015); (ii) Beck et al. (2010a); (iii) Leung and Jiao (2006); (iv) WHO (1989); (v) Niencheski and Windom (2015); (vi) O’Connor et al. (2022)
	Chromium (Cr)	Aquifer lithology (i)	Metal processing (ii); tanneries (ii); cement, automobile, and pigment production (ii); chemical processing (ii); wastewater (iii–v); agriculture (ii, v)	Redox conditions (iii, vi–vii); anthropogenic inputs (iii, v)	(i) McClain et al. (2019); (ii) Saha et al. (2011); (iii) Szymczycha et al. (2016); (iv) Leung and Jiao (2006); (v) Prakash et al. (2021); (vi) O’Connor et al. (2015); (vii) O’Connor et al. (2022)
	Manganese (Mn)	Aquifer lithology (i); advection from reducing sediments (ii)	Wastewater (iii); steel production (iv); batteries (iv); metal alloys (iv); fungicides (iv); pigments (iv)	Physical mixing (v); redox conditions (vi–ix); organic carbon remineralization (x)	(i) Zhang et al. (2020); (ii) Beck et al. (2008); (iii) Leung and Jiao (2006); (iv) Bradl (2005); (v) Liu et al. (2019b); (vi) Niencheski and Windom (2015); (vii) Beck et al. (2010a); (viii) Beck et al. (2007); (ix) Beck et al. (2009); (x) Aller et al. (1996)

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**Table 1.** Continued

Metal group	Metal	Main coastal source(s)—natural	Main coastal source(s)—anthropogenic	Applications in SGD work	References
	Iron (Fe)	Aquifer lithology (i)	Mining (ii); wastewater (iii); construction (iv); metal production (iv)	Redox conditions (v–viii); organic carbon remineralization (ix)	(i) Zhang et al. (2020); (ii) Alorda-Kleinglass et al. (2019); (iii) Leung and Jiao (2006); (iv) Bradl (2005); (v) Niencheski and Windom (2015); (vi) Kim and Kim (2015); (vii) Sanders et al. (2017); (viii) Bone et al. (2006); (ix) Aller et al. (1996)
	Cobalt (Co)	Aquifer lithology (i)	Mining (ii); metal and glass production (iii); paint (iii); wastewater (iv)	Redox conditions (v); anthropogenic inputs (ii, iv–ix)	(i) Jeong et al. (2012); (ii) Alorda-Kleinglass et al. (2019); (iii) Bradl (2005); (iv) Beck et al. (2009); (v) Trezzi et al. (2016); (vi) Tovar-Sánchez et al. (2014); (vii) Szymczycha et al. (2016); (viii) Samanta and Dalai (2018); (ix) Tamborski et al. (2020a)
	Nickel (Ni)	Aquifer lithology (i); weathering (i)	Mining (ii); metal alloys (iii); batteries (iii); pigments (iii); electronics (iii); agriculture (i); landfills (i); wastewater (vi–v)	Redox conditions (ii; vi); anthropogenic inputs (i–ii; iv–ix)	(i) Prakash et al. (2021); (ii) Trezzi et al. (2016); (iii) Bradl (2005); (iv) Beck et al. (2009); (v) Tamborski et al. (2020a); (vi) Jeong et al. (2012); (vii) Tovar-Sánchez et al. (2014); (viii) Samanta and Dalai (2018); (ix) Luo et al. (2022)
	Copper (Cu)	Aquifer lithology (i); mineral ores (i)	Mining (ii); conductors (i); plumbing (i); antifouling boat paint (iii–iv); industrial waste (i); agriculture (v); wastewater (vi)	Redox conditions (ii); anthropogenic inputs (ii–xi)	(i) Bradl (2005); (ii) Trezzi et al. (2016); (iii) Charette and Buesseler (2004); (iv) Tamborski et al. (2020a); (v) Prakash et al. (2021); (vi) Beck et al. (2009); (vii) Jeong et al. (2012);

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**Table 1.** Continued

Metal group	Metal	Main coastal source(s)—natural	Main coastal source(s)—anthropogenic	Applications in SGD work	References
					(viii) Tovar-Sánchez et al. (2014); (ix) Waska et al. (2019); (x) Samanta and Dalai (2018); (xi) Luo et al. (2022)
	Zinc (Zn)	Aquifer lithology (i); mineral ores (i)	Mining (ii–iii); metal alloys (i); batteries (i); PVC (i); electronics (i); medicine and chemical production (i); antifouling boat paint (iv–v); agriculture (vi); wastewater (v, vii)	Redox conditions (v–vi); anthropogenic inputs (ii–xii)	(i) Bradl (2005); (ii) Alorda-Kleinglass et al. (2019); (iii) Trezzi et al. (2016); (iv) Charette and Buesseler (2004); (v) Tamborski et al. (2020a); (vi) Prakash et al. (2021); (vii) Beck et al. (2009); (viii) Tovar-Sánchez et al. (2014); (ix) Wang et al. (2019); (x) Szymczycha et al. (2016); (xi) Luo et al. (2022); (xii) Bhagat et al. (2022)
	Molybdenum (Mo)	Aquifer lithology (i); seawater (i)	Mining (ii); metal alloys (ii); electronics (ii); industrial wastewater (ii)	Redox conditions (iii); anthropogenic inputs (iv)	(i) Dalai et al. (2005); (ii) Bradl (2005); (iii) Prakash et al. (2021); (iv) Tovar-Sánchez et al. (2014)
	Silver (Ag)	Aquifer lithology (i); mineral ores (i–ii)	Mining (i–ii); industry (i); wastewater (iii)	Anthropogenic inputs (i–iii)	(i) Rivera-Duarte and Flegal (1997); (ii) Alorda-Kleinglass et al. (2019); (iii) Beck et al. (2009)
	Cadmium (Cd)	Aquifer lithology (i); weathering (i); wildfires (i), sea salt (i); volcanoes (i)	Mining (ii–iii); metal industry (iv); metal alloys (i, iv); batteries (iv); pigment production (iv); agriculture (iv); fertilizers (iv); wastewater (i); fossil fuel combustion (i, iv)	Anthropogenic inputs (ii–vii)	(i) Kubier et al. (2019); (ii) Trezzi et al. (2016); (iii) Alorda-Kleinglass et al. (2019); (iv) Bradl (2005); (v) Szymczycha et al. (2016); (vi) Tovar-Sánchez et al. (2014); (vii) Wang et al. (2019)
	Rhenium (Re)	Aquifer lithology (i)	Metal alloys (i–ii); mining (i–ii)	Anthropogenic inputs (i–ii)	(i) Leybourne and Cameron (2008); (ii) Ketterer (1990)

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**Table 1.** Continued

Metal group	Metal	Main coastal source(s)—natural	Main coastal source(s)—anthropogenic	Applications in SGD work	References
Post-transitional	Mercury (Hg)	Aquifer lithology (i); volcanoes (ii)	Fossil fuel combustion (iii), electrical products (iv), industry (iv), fungicides (iv), pharmaceutical preservative (iv), metal solvents (iv); wastewater (v)	Aquifer water mixing (i, vi–vii), anthropogenic inputs (iii, v–vi)	(i) Bone et al. (2007); (ii) Ganguli et al. (2014); (iii) Wang et al. (2019); (iv) Bradl (2005); (v) Szymczycha et al. (2013); (vi) Ci et al. (2023); (vii) Black et al. (2009)
	Aluminum (Al)	Aquifer lithology (i); weathering (i–ii); acid sulfate soils (iii)	Mining (iv); metal alloys (iv); industry (iv); wastewater (iv)	Chemical weathering (i–ii); seawater–freshwater interactions (i–ii); redox conditions (i–iii); anthropogenic inputs (iv)	(i) Prakash et al. (2021); (ii) Beck et al. (2007); (iii) Santos et al. (2011); (iv) Botté et al. (2022)
	Thallium (Tl)	Groundwater (i); aquifer lithology (i)	Mining (ii); metal alloys (ii); electronics (iii); optics (iii)	Anthropogenic inputs (ii); redox conditions (i)	(i) Ahrens et al. (2020); (ii) Bradl (2005)
	Lead (Pb)	Aquifer lithology (i); mineral ores (ii); soils (iii)	Mining (iv–v) batteries (ii); ammunition (ii); metal alloys (ii); electronics (ii); pigments (ii); fertilizers (iii–vi); wastewater (i, iii)	Anthropogenic inputs (i, iii–v); redox conditions (vi)	(i) Beck et al. (2007); (ii) Bradl (2005); (iii) Prakash et al. (2021); (iv) Alorda-Kleinglass et al. (2019); (v) Trezzi et al. (2016); (vi) Qu et al. (2020)
Metalloids	Boron (B)	Precipitation; aquifer lithology (i)	Detergents; fertilizers; wastewater (i, iii)	Anthropogenic inputs (i–iii)	(i) Barth (1998); (ii) Tamborski et al. (2020b); (iii) Vengosh et al. (1999)
	Silica (Si)	Aquifer lithology (i–iii); biogenic silica (i–iii)	—	Groundwater tracer (ii–iii)	(i) Kim et al. (2005); (ii) Rahman et al. (2019); (iii) Oehler et al. (2019)
	Arsenic (As)	Aquifer lithology (i–iv)	Mining (v); glass and textile production (v); pigments (v); fireworks (v); preservatives (v); animal feed (v); industrial waste (ii, vi–vii)	Anthropogenic inputs (i–iv; vi–vii); redox conditions (viii)	(i) Berg et al. (2001); (ii) Chen and Liu (2007); (iii) Shaji et al. (2021); (iv) Smedley and Kinniburgh (2013); (v) Bradl (2005); (vi) Luo et al. (2022); (vii) Sun et al. (2017); (viii) Bone et al. (2006)
	Antimony (Sb)	Mineral deposits (i–iii)	Mining (i–iii); metal alloys (i); industry		(i) Ilgen et al. (2014); (ii) Fang et al.

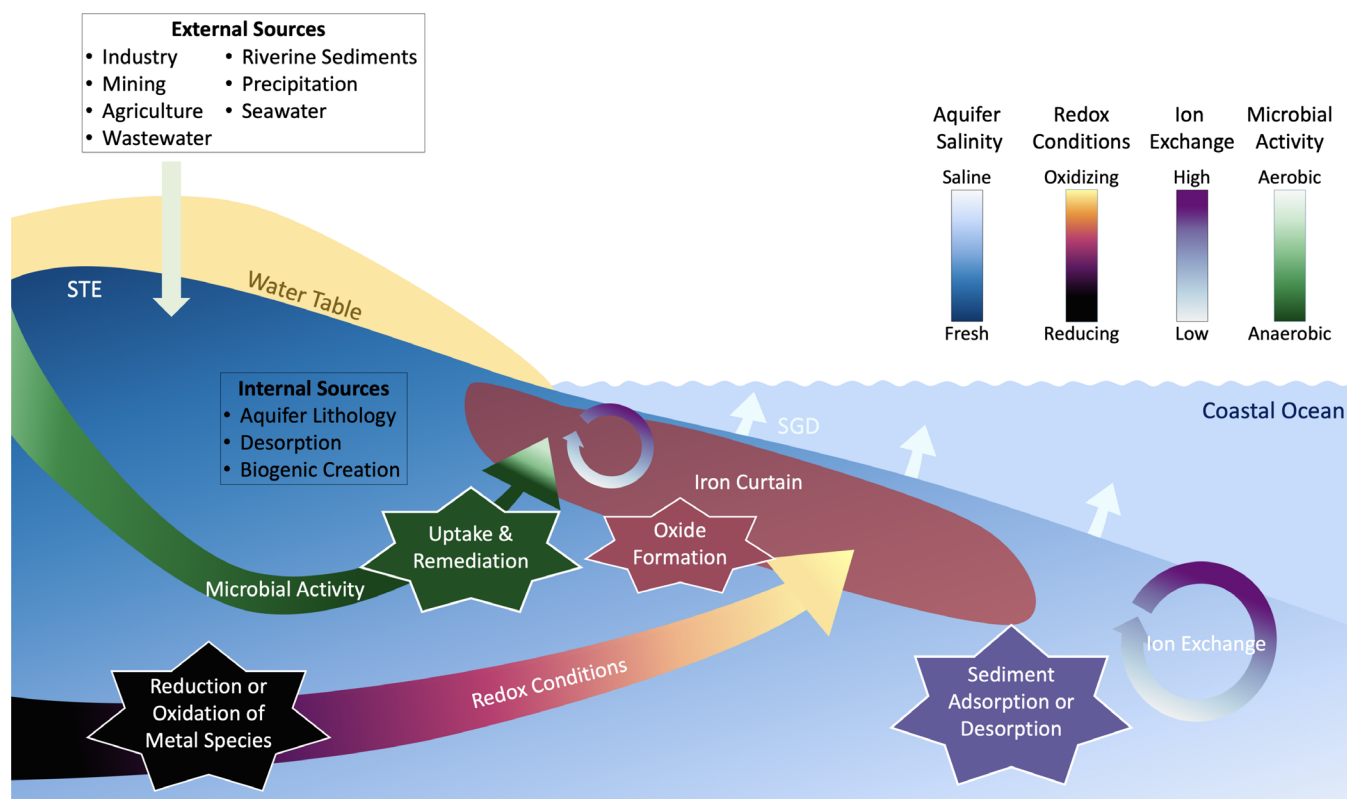
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**Table 1.** Continued

Metal group	Metal	Main coastal source(s)—natural	Main coastal source(s)—anthropogenic	Applications in SGD work	References
			(i–iii); ammunition (i)	Anthropogenic inputs (i–iii); redox conditions (i–iii)	(2021); (iii) Wilson et al. (2004)
REEs	Yttrium (Y)	Mineral ores (i)	Mining (i); electronics (i)	Groundwater–wastewater (i) interactions; anthropogenic inputs (i)	(i) Cendón et al. (2022)
	LREEs: Lanthanum (La), cerium (Ce), praseodymium (Pr), neodymium (Nd)	Aquifer lithology (i); mineral ores (ii); weathering (i)	Mining (ii); electronics (ii); fertilizers (ii); animal feed (ii)	Seawater–freshwater interactions (i, iii–vi); redox conditions (i, iii–vi); chemical weathering (i); anthropogenic inputs	(i) Johannesson et al. (2017); (ii) Piarulli et al. (2021); (iii) Chevis et al. (2015); (iv) Duncan and Shaw (2003); (v) Kim and Kim (2011); (vi) Liu et al. (2016)
	MREEs: Samarium (Sm), europium (Eu), gadolinium (Gd), terbium (Tb), dysprosium (Dy), holmium (Ho)	Aquifer lithology (i); mineral ores (ii); weathering (i)	Mining (ii); electronics (ii); MRI contrast (Gd; i–iii)	Seawater–freshwater interactions (i, iii–vii); redox conditions (i, iii–vii); chemical weathering (i); anthropogenic inputs (i, iii); wastewater tracer (i, iii)	(i) Johannesson et al. (2017); (ii) Piarulli et al. (2021); (iii) Paffrath et al. (2020); (iv) Chevis et al. (2015); (v) Duncan and Shaw (2003); (vi) Kim and Kim (2011); (vii) Liu et al. (2016)
	HREEs: erbium (Er), thulium (Tm), ytterbium (Yb), lutetium (Lu)	Aquifer lithology (i); mineral ores (ii); weathering (i)	Mining (ii); electronics (ii)	Seawater–freshwater interactions (i, iii–vi); redox conditions (i, iii–vi); chemical weathering (i); anthropogenic inputs	(i) Johannesson et al. (2017); (ii) Piarulli et al. (2021); (iii) Chevis et al. (2015); (iv) Duncan and Shaw (2003); (v) Kim and Kim (2011); (vi) Liu et al. (2016)
	Uranium (U)	Aquifer lithology (i–iii); weathering (ii); riverine sediments (i)	Nuclear fission product (i); mining (i); fertilizers (iii); wastewater (iv–v)	Redox conditions (iii); aquifer source (i); seawater–freshwater interactions (iii); chemical and physical weathering (i); groundwater tracer (i–iii)	(i) Charette et al. (2008); (ii) Wang and You (2013); (iii) Santos et al. (2011); (iv) Prakash et al. (2021); (v) Windom and Niencheski (2003)
	Thorium (Th)	Aquifer lithology (i); riverine sediments (i); weathering (ii)	Fertilizers (iii); industry (iii)	Adsorption/release on Fe/Mn oxides (i); anthropogenic inputs (iii)	(i) Charette et al. (2008); (ii) Swarzenski (2007); (iii) Prakash et al. (2021)





**Fig. 1.** Major processes influencing metal behavior in the STE.

for elevated metal concentrations (e.g., V, Cr, Mn, Co, Ni, Cd; Chen and Jiao 2008), where previously sediment-bound metals can be mobilized by increasing salinity (Adyari et al. 2021). Continuous contact with higher salinities resulted in  $^{137}\text{Cs}$  fluxes 15 times greater via porewater diffusion compared to river discharge off the coast of Japan due to desorption upon contact with saltwater ions (Kambayashi et al. 2021). Sea-level rise and seawater intrusion may also sharpen the fresh-saline interface, offering less opportunity for metal filtration and bioremediation prior to discharge to the coastal ocean.

#### Precipitation and storm events

While climate change-driven precipitation changes will vary substantially by region, increases in heavy rainfall are expected for most areas globally whereas mean annual rainfall changes (increase/decrease) are region specific (Richardson et al. 2024). Precipitation changes can alter the hydraulic gradient, aquifer storage and recharge, and SGD fluxes (Richardson et al. 2024). For instance, in areas where rainfall decreases, the combination of lower aquifer levels and sea-level rise could reduce SGD volume, while simultaneously leading to increased releases of metals to the STE and coastal ocean via SGD due to increased contact with saline water (Roy et al. 2010). El Niño Southern Oscillation precipitation

variations have been linked to shifts in recharge rates, where a reduced fresh SGD seepage face area resulted in lower Fe fluxes, while Fe reduction rates remained relatively stable (Roy et al. 2013).

Storm events (e.g., extreme precipitation, tropical cyclones), which include processes such as flooding, storm surges, big waves, physical pressure, and disruption of the hydraulic and redox gradient, can lead to an acute release of metals to the coastal ocean. Storm-driven waves were associated with the rapid release of redox-sensitive metals including As, Fe, and Mn (Rakhimbekova et al. 2018). Storm surges and associated seawater intrusion during Hurricane Sandy drove release of Pb, As, Cu, Cr, and Fe from the Raritan Bay Slag Superfund site to the coast (Personna et al. 2014), highlighting the vulnerability of coastal areas to legacy contamination. Storm events can also lead to increased surficial flow (flooding, riverine discharge, overland flow) of particulate-bound metals to the coast (Takata et al. 2020).

#### Warming

Both atmospheric and oceanic warming impact coastal aquifer and ocean temperatures, decreasing aquifer oxygen concentrations, and modifying redox conditions and microbial communities (Knee and Paytan 2011; Richardson et al. 2024). Increasing temperatures speed up microbial

reactions, impacting the many metals susceptible to microbial reduction (e.g., Fe, Mn, Se, Cr, Hg, Tc, V, Mo, Cu, Au, Ag, U; Lovley 1993; Knee and Paytan 2011; Santos et al. 2011; O'Connor et al. 2015). For example, warming led to increased metabolism of bacteria leading to Hg methylation and increasing the Hg concentration in solution (Knee and Paytan 2011). There is also evidence that increases in coastal temperature may drive changes in salinity and hydraulic gradients, changing the redox conditions in the STE and possibly leading to increases in SGD (Nguyen et al. 2020).

### *Coastal acidification and pH changes*

Decreases in STE and coastal ocean pH, driven by land, oceanic, and atmospheric processes can increase desorption of metals (e.g., Cu, As, REEs; Hu et al. 2006; Zeng et al. 2015). Decreasing pH may lead to environmental conditions that favor metals such as Cu(I) and As(III) in their reduced and more toxic forms (Zeng et al. 2015). Model-predicted impact of decreasing pH and increasing water temperature led to increasing estuarine  $\text{Cu}^{2+}$  by 115% in the next 100 yr (Richards et al. 2011). Metals such as Hg and Pb can also synergistically decrease the efficacy of photosynthesis, enhancing ocean acidification and favoring conditions that further favor metal bioavailability (Zeng et al. 2015). Some metals (e.g., Cd, Ni) can follow a diel cycle due to pH-dependent changes associated with photosynthesis-respiration, with adsorption during light periods corresponding to pH increases, and slower and incomplete desorption during dark periods (Beck et al. 2010b). In some cases, acid rain has also been associated with STE pH decreases, leading to enhanced mobilization of metals in the STE (e.g., Cd, Co, Ni; Kj  ller et al. 2004).

Metals are also more readily released from solid and colloidal phases in acidic STE conditions, such as acid sulfate soil systems (Santos et al. 2011). Groundwater was the main pathway for Al release to a local estuary following Al dissolution under extreme low pH conditions in an acid sulfate soil system (Santos et al. 2011). High rainfall events resulting in flooding of acid sulfate soil systems can also decrease aquifer pH, resulting in increased discharge of U and Ba to the coastal ocean (Moody et al. 2022). Increasing pH from seawater intrusion can also lead to mobilization of Al, Fe, Ni, Mn, and Zn from acid sulfate soils (Wong et al. 2010).

### *Simultaneous influence on redox conditions from climate change hazards*

Sea-level rise, precipitation changes, storm events, warming atmospheric and ocean temperatures, and ocean acidification collectively impact STE redox conditions and thus metal speciation. Changes in redox conditions are driven by the intrusion of oxygen rich seawater mixing with anoxic groundwater, temperature shifts, organic matter composition, microbial communities present, and electron acceptor availability (O'Connor et al. 2018; Goyetche et al. 2022; Richardson et al. 2024).

Climate change may result in competing processes influencing STE redox conditions and metal speciation. For instance, increasing temperatures decrease aquifer oxygen levels, while influxes of oxygenated seawater may increase STE oxygen concentrations. While it may be difficult to predict or isolate simultaneous deviations from baseline conditions, case studies can provide valuable information about metal behavior under a changing climate. For example, seasonal shifts from reducing to oxidizing STE conditions can transform intertidal sediments from sinks to sources of certain metals (e.g., U, Tl; Charette and Sholkovitz 2006; Ahrens et al. 2020; Reckhardt et al. 2015; Sanders et al. 2017; Prakash et al. 2021). Intertidal sediments can then become sources of Tl, U, and Re due to reoxidation during winter (Ahrens et al. 2020). Reducing conditions can result in the STE acting both as a metal sink (through metal oxide formation and scavenging) and source (through Fe- and Mn-oxide dissolution). The iron curtain, a zone of Fe-oxide precipitation, plays a critical role in many STEs (Charette and Sholkovitz 2002). Fe-oxides have a large sorption capacity, effectively trapping phosphorus (P), dissolved organic matter (DOM), and some metals at the iron curtain (Charette and Sholkovitz 2002, 2006; Beck et al. 2010a).

Metal oxide formation and scavenging under reducing conditions can remove some metals from solution. For instance, lower STE Cr concentrations were observed in a reducing STE due to precipitation of highly mobile Cr(VI) to readily adsorbed Cr(III) species (Szymczycha et al. 2016). In another example, Cu mineral precipitation (e.g., cuprite) following Cu reduction by dissolved Fe(II) was hypothesized as an explanation for Cu removal coinciding with dissolved Fe removal in a sulfate-poor, freshwater region of a shallow aquifer (Beck et al. 2010a). Sulfate reduction is an important process in highly reducing STEs and can cause sulfide precipitation and adsorption of metals (e.g., Pb, REEs) onto particles (Qu et al. 2020; Paffrath et al. 2020). Pb scavenging also occurs through association with colloidal Al, removing both along the aquifer flow path (Beck et al. 2007).

Fe- and Mn-oxide dissolution under reducing conditions can also lead to co-release of metals bound to the coastal aquifer matrix to solution. For instance, Fe/Mn (hydro)-oxide dissolution was associated with increased dissolved As, U, and Mo concentrations under reducing conditions (Bone et al. 2006; Beck et al. 2007, 2009, 2010a; Smedley and Kinniburgh 2013; Sanders et al. 2017). Similarly, Cd, Ni, and Zn exhibit similar patterns to dissolved Fe and Mn in groundwater, suggesting control by dissolution/precipitation of ferromanganese solids (Niencheski and Windom 2015).

Changes in the presence, composition, and quality of DOM can directly influence the transferring efficiency of a metal across the sediment water interface. Groundwater systems are typically low oxygen environments and rich in dissolved organic carbon (DOC; includes humic and non-humic material) compared to surface waters (Knee and Paytan 2011).

Furthermore, many metals are suspended in colloidal form in groundwater, particularly those that bind with humics (Knee and Paytan 2011), but incursions of labile marine organic matter can result in increased metals in solution (Richardson et al. 2024). DOM plays a crucial role in binding transition metals such as Cu, Ni, Co, Zn, V, and Cr (Beck et al. 2007, 2009, 2010a; O'Connor et al. 2015, 2022) in the dissolved form and Mn and Pb as colloids (Kim and Kim 2015). High DOM concentrations can cause STE conditions that favor attenuation of metals from solution, resulting in relatively immobile phases such as metallic sulfides (O'Connor et al. 2015). DOC may help mobilize metals such as Ni, Hg, As, Cr, and Cu (Christensen et al. 1996; Kalbitz and Wennrich 1998) and in some cases, Cd and Zn (Christensen et al. 1996). Seasonal fluctuations in DOC can have been linked to variations in Fe, Mn, V, and Cr concentrations in STEs (O'Connor et al. 2015, 2018, 2022).

### Metals as tracers of climate change

Metals can be used to trace water sources and transport, providing important information about groundwater evolution, aquifer source, and fresh-saline water interactions (Table 1). In the context of anthropogenic impact, these applications can be extended to better understand sources of contaminated water bodies, surface vs. groundwater transport pathways, and potential effects from sea-level rise such as aquifer salinization. Future work monitoring metal concentrations over time may help address knowledge gaps regarding how SGD fluxes and resulting metal discharge will shift due to climate change.

Alkali and alkali earth metals that are associated with salts, such as K, Na, Ca, and Mg, can be used to better understand fresh-saline water interactions. For instance, Schiavo et al. (2006) compared K and nitrate concentrations for fresh and saline water masses to explain seawater mixing. In another example,  $^{40}\text{K}$  ( $t_{1/2} = 1600$  yr) and  $^{222}\text{Rn}$  were used to separate the influence of precipitation from seawater (Patiris et al. 2021). Na vs. Cl ratios can be used to differentiate between seawater intrusion and evaporite dissolution (Nouayti et al. 2022; Salem et al. 2016). Similarly, Ca and Mg vs. Cl ratios can be used to separate meteoric and oceanic water sources (Tillman et al. 2014).

Other metals, such as Sr and Mn, can also trace fresh-saline water interactions in the coastal aquifer (Musgrove and Banner 1993; Jørgensen et al. 2008; Russak et al. 2016). In most STEs, Sr concentrations are conservative, but dissolved Sr in seawater exchanging with brackish groundwater and aquifer-sorbed Sr, can result in nonconservative  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios in the STE (Beck et al. 2013). However, nonconservative mixing of Ca and Sr concentrations and isotopes in the STE is an important consideration when using Sr isotopes, particularly in carbonate environments (Holmden et al. 2012). Similarly, laboratory experiments indicate that Mn may be able to differentiate between salinization and freshening events in the

fresh-saline water interface zone of coastal aquifers, since  $\text{Mn}^{2+}$  more likely to be released to porewater during salinization and depleted during freshening due to its strong affinity with the solid phase (Russak et al. 2016).

Elevated concentrations of some metals in the STE can also be used to understand groundwater evolution and aquifer source. Through the lens of anthropogenic change, this is useful for differentiating water sources in a multiple aquifer system where contaminant sources may differ. For instance, alkali and alkali earth metals such as Li (particularly Li stable isotopes) and Ba can be useful for understanding groundwater evolution and source, and chemical weathering processes (e.g., Edmond et al. 1985; Santos et al. 2011; Bagard et al. 2015; Martin et al. 2020; Mayfield et al. 2021). Other metals, such as Nd, can provide information on the source and age of the groundwater (e.g., Elderfield and Greaves 1982; van de Flierdt et al. 2016).

In the context of SGD, metals such as Ra, Si, Sr, and Ba have been applied as groundwater tracers (e.g., Oehler et al. 2019; Garcia-Orellana et al. 2021). Ra is the most well-known metal tracer of groundwater (e.g., Garcia-Orellana et al. 2021). It has wide applications for tracing groundwater discharge and water residence times as discussed in Garcia-Orellana et al. (2021). Ba can act as a stable analog for Ra as it typically behaves similarly and has applications as an SGD proxy in coastal systems not dominated by river inputs (Moore 1997; Moore and Shaw 2008). More recently, Sr concentrations and slightly depleted relative to seawater radiogenic  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios have been used to estimate SGD fluxes in the Bengal Basin, which is a significant source to the global seawater Sr budget (Beck et al. 2013; Chakrabarti et al. 2018).

### Human pressure on metal sources to the coastal ocean

Anthropogenic escalation of metal sources is putting increasing pressure on coastal systems. Metal concentrations in coastal waters (e.g., Zn, Pb, Cd, Cu, As) are 5–10 times greater compared to the 1<sup>st</sup> half of the 20<sup>th</sup> century (Mashiatullah et al. 2012; Murkherjee et al. 2024). Increasing coastal population and urbanization, wastewater, agricultural, and industrial production represent an increasing threat of metal pollution to the coastal ocean. Legacy contamination remaining in coastal sediments and landfills are not only generally poorly documented (Nicholls et al. 2021), but also represent an important source of metals (e.g., metal scraps, solid wastes, electronic waste, batteries). Compounding effects from climate change, such as sea-level rise and storm surges further threaten to expose and mobilize metals in coastal landfills, wastewater infrastructure, urban areas, agricultural sites, mines, and legacy deposits.

Sewage and domestic wastewater can be a substantial source of metals (e.g., Zn, Cr, Cu, Cd, Fe, Mn, V, Co, REEs) to the environment (Leung and Jiao 2006; Kulaksız and Bau 2007; Paffrath et al. 2020). An estimated 56% of domestic wastewater is treated globally (United Nations [UN] Habitat

and World Health Organization [WHO] 2021). For contaminated sites, SGD-derived fluxes of As, Cd, Cu, Ni, and Zn can be the same order of magnitude if not greater than river-derived fluxes (Luo et al. 2022). Wastewater leakage into groundwater is typically characterized by low pH and low oxygen concentrations. Wastewater also typically contains high DOM concentrations, leading to significant increases in Cu, Ni, and Zn solubilities in the STE due to complexation with organic ligands (Beck et al. 2009). Beyond the potential for SGD to be a direct source of wastewater-derived metals to the coastal ocean, some metals may be used for source tracking. For instance, Tamborski et al. (2020b) used B isotopes to trace nitrogen (N) sources in the coastal aquifer. B is added to aquifer systems both naturally (natural precipitation) and anthropogenically (detergents, fertilizers, sewage, septic systems) (Barth 1998; Vengosh et al. 1999; Shireen et al. 2018).

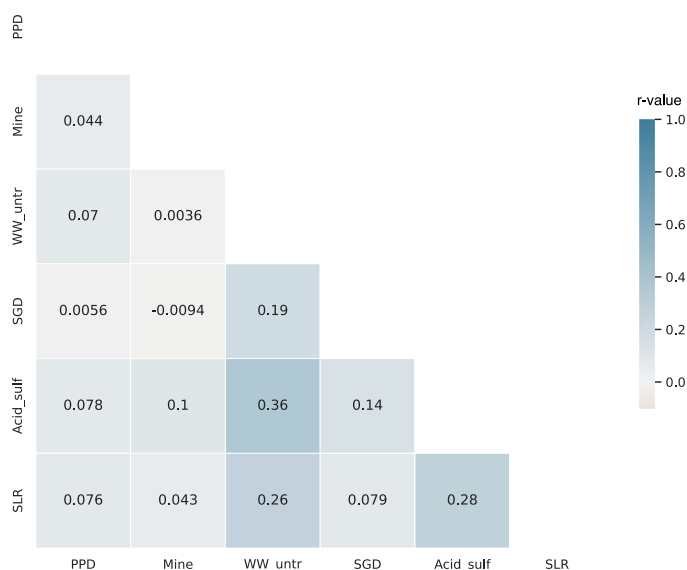
Wastewater from hospitals can also be a major source of metals such as Gd (contrast agent) and Hg (diagnostic agents, disinfectants, diuretics) (Bau and Dulski 1996; Verlicchi et al. 2010). Hospital wastewater typically contains significantly higher concentrations of metals, emerging contaminants, nitrogen, and biochemical oxygen demand, compared to domestic sewage (Verlicchi et al. 2015). Because hospital wastewater is typically processed through municipal wastewater treatment plants (WWTP; provided wastewater treatment is available), it may not fully target or address the demand of the high concentrations present (Verlicchi et al. 2015). Gd associated with hospital wastewater has been detected in SGD

and coastal areas and has been suggested as a possible tracer for anthropogenic influence (Kulaksız and Bau 2007; Hatje et al. 2016; Johannesson et al. 2017; Andrade et al. 2020).

Agriculture (e.g., fertilizers, pesticides) and industry (e.g., industrial byproducts, paints) can be sources of Pb, Cu, Cr, Ni, Cd, Zn, and REEs to the aqueous environment (Noyes et al. 2009; Trezzi et al. 2016; Liu et al. 2019a; Prakash et al. 2021; Luo et al. 2022). For instance, excess concentrations of Ce and Eu at a farm in Jiangxi Province, China were attributed to application of phosphate fertilizers containing high levels of REEs (Liu et al. 2019a). Excess phosphates have also been linked to remobilization of As by competing with arsenate for the adsorption site on Fe and Mn hydroxides (Sun et al. 2017; Pan et al. 2019; Luo et al. 2022). In the coastal zone, harbor operations and antifouling paints can be a major source of metals such as Cu, Zn, and Pb (Charette and Buessler 2004; Rodellas et al. 2014). Antifouling paints frequently contain high concentrations of Cu; while direct surface inputs of Cu from antifouling paints were determined to be the main source of Cu to the Elizabeth River Estuary, USA, SGD contributed  $\sim 3\%$  of Cu to the system (Charette and Buessler 2004).

Mining operations are rapidly expanding, with global production increasing by 52% between 2000 and 2020 (Reichl and Schatz 2022). Production of metals such as Li, Ga, and REEs have seen the largest increases over that time span, increasing by 582%, 485%, and 176%, respectively (Reichl and Schatz 2022). While limited global data exist (representing only 4% of global population), only 30% of industrial wastewater (including mining, construction, manufacturing, electricity production and distribution) are treated (UN Habitat and WHO 2021). Mining operations can deliver large quantities of metals such as Cu, Pb, Fe, Zn, Au, Ag, Hg, and REEs to the coastal ocean via SGD (Trezzi et al. 2016; Balaram 2019; Alorda-Kleinglass et al. 2019; Adyasari et al. 2021). For example, Au and Ag mining tailings, which can include other metals such as Hg, can be present in concentrations well above health regulatory thresholds along the coast (Adyasari et al. 2021). Increasing demand for REEs has resulted in environmental degradation associated with large-scale dumping of electronic waste as well as significant human health issues from mining (Balaram 2019).

Excess metal inputs can lead to substantial declines in water quality and can aggravate other processes such as eutrophication. For instance, metals such as B, Co, Cu, Fe, and Mo can further drive cyanobacteria growth under eutrophic conditions (Zhang 2000; Downs et al. 2008). Metals can also directly interact with N cycling; for instance, excess Cd concentrations enhanced N fixation through formation of cadmium sulfide production in Baltic Sea sediments (Broman et al. 2019). Thus, considering metal interactions with other elemental cycles is critical in the context of global change. For instance, global anthropogenic inputs of N have doubled



**Fig. 2.** Correlation (Pearson-score  $r$ -values) between normalized model input variables: population density (PPD), presence/absence of mines (Mine), wastewater treatment level (WW\_untr), SGD, presence/absence of acid sulfate soils (Acid\_sulf), sea-level rise (SLR). Low  $r$ -values between input variables indicate independence between model inputs.



**Table 2.** Classification values assigned for each dataset in the metal risk model.

Variable	Range	Value assigned	Description
Population density (people km <sup>-2</sup> )	< 50	0	Weak human pressure
	50–200	0.25	Moderate human pressure
	200–1000	0.50	High human pressure
	1000–5000	0.75	Very high human pressure
	> 5000	1.0	Extreme human pressure
Untreated wastewater (%)	0	0	No untreated wastewater input
	1–25	0.25	Low untreated wastewater input
	26–50	0.50	Moderate untreated wastewater input
	51–75	0.75	High untreated wastewater input
	> 76	1.0	Very high untreated wastewater input
Mines	0	0	Absent
	1	1.0	Present
Sea-level rise (cm)	–200 to 0	0	Sea-level decrease
	0–300	0.25	Low sea-level rise
	300–600	0.50	Moderate sea-level rise
	600–900	0.75	High sea-level rise
	> 900	1.0	Very high sea-level rise
SGD (m <sup>2</sup> yr <sup>-1</sup> )	< 2	0	None to very low SGD
	2–10	0.25	Low SGD
	10–320	0.50	Moderate SGD
	320–800	0.75	High SGD
	> 800	1.0	SGD hotspot
Acid sulfate soils	0	0	Absent
	1	1.0	Present

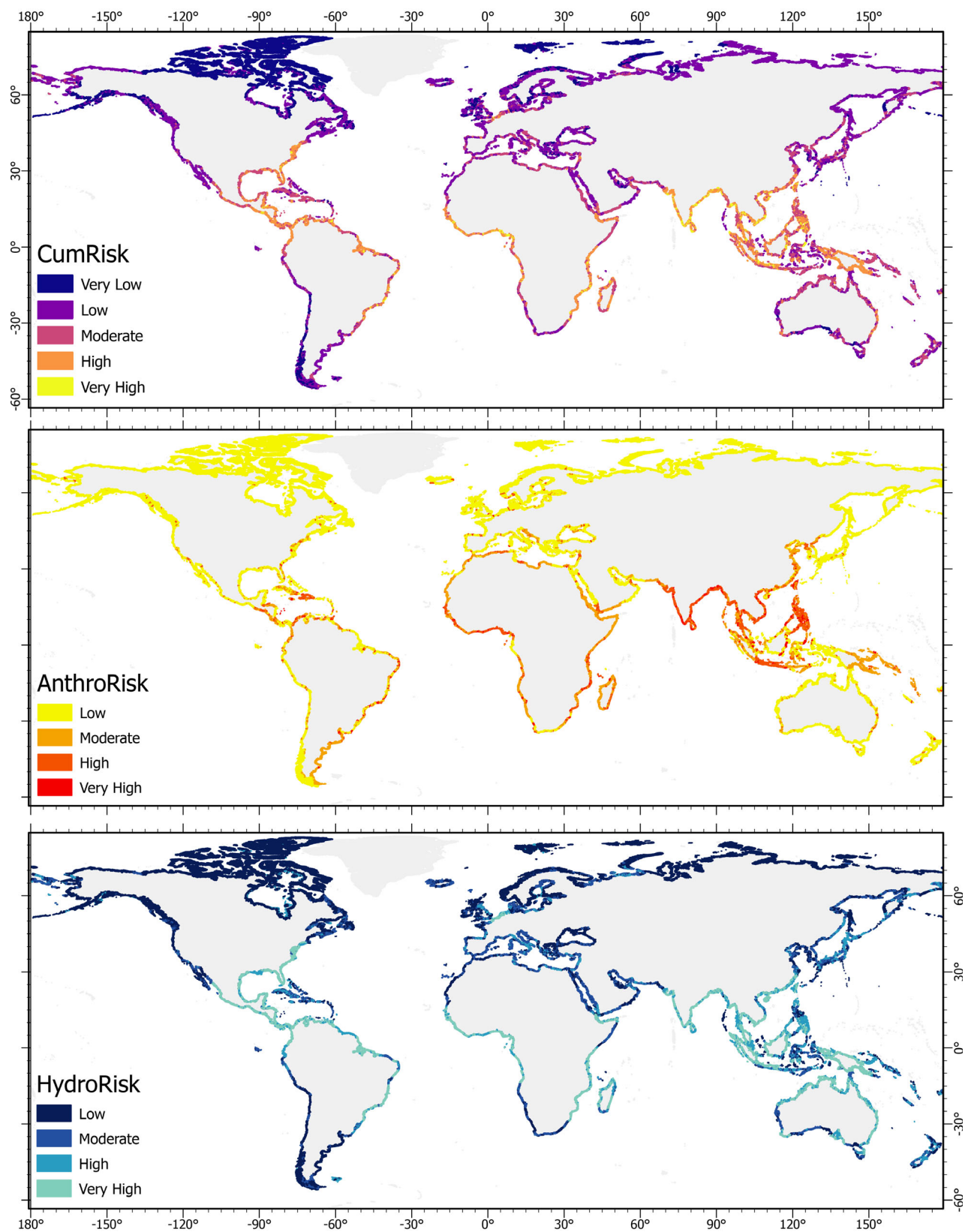
since 1950 and are projected to double again by 2050 (Vitousek et al. 1997; Jickells et al. 2017), where metal inputs may further exacerbate for instance, eutrophication.

Beyond declining water quality, SGD can be a major vector for metals toxic to marine life and humans (e.g., Laurier et al. 2007). Hg, Cr(VI), Cd, Cu, Pb, Zn, As, and Ni are specifically concerning due to their ability to bioaccumulate and toxicity to both marine life and humans (e.g., Adyasari et al. 2021). Locations with higher SGD have been linked to not only increased concentrations of metals such as Hg in the water column and sediments, but also in marine life such as mussels (Laurier et al. 2007).

### Global risk assessment

To evaluate the risk for SGD to deliver metals to the coastal ocean, we developed a global hotspot risk model. Coastal watersheds were defined from the BasinATLAS dataset (15 arc-second resolution; Linke et al. 2019). Sources of anthropogenic pressure included population density (Linke et al. 2019), wastewater treatment level (UN Data 2023), and presence of mines (10 km buffer, Orris and Grauch 2002;

Jasansky et al. 2022, 2023). Hydrogeological pressure sources considered in the model included sea-level rise projections (CMIP6, SSPs 8.5, 2° warming with respect to 1850–1900; Iturbide et al. 2021; Gutiérrez et al. 2021), SGD (Luijendijk et al. 2020), and presence of acid sulfate soils (Proske et al. 2014). Model input variables were selected based on availability of global data, influence on SGD metal concentrations, and independence between input variables (Fig. 2). Wastewater treatment data (UN Data 2023) were spatially matched first by country (GADM 2022) and then to each coastal watershed. All values were min–max normalized and classified by quintile values to allow for comparison between variables, except for population density, which was classified using literature values (Table 2; Food and Agriculture Organization [FAO] 2016). Given uncertainties associated with the importance of an individual stressor, all risk variables were assigned the same weight. These values were used to generate cumulative risk (CumRisk) by summing all risk variables following a similar approach to Halpern et al. 2008. Model uncertainty was by conducting jack-knife sensitivity testing (Jones et al. 2018). Briefly, this method accounts for the effect of each variable on the cumulative impact, resulting in an  $r^2$



**Fig. 3.** Risk of metal release and pollution to the coastal ocean via SGD. CumRisk represents the relative cumulative risk from both anthropogenic (AnthroRisk) and hydrogeological (HydroRisk) factors.

**Table 3.** Mean  $\pm$  standard deviation values resulting from the risk analysis. Values in parentheses are the resulting  $r^2$  values from the sensitivity analysis, where lower  $r^2$  values indicate greater influence on the final CumRisk.

CumRisk group and threshold	Anthropogenic pressure			Hydrogeological pressure		
	Pop. density (people km <sup>-2</sup> )	Mining (%)	Untreated WW (%)	SLR (cm)	SGD (m <sup>2</sup> yr <sup>-1</sup> )	Acid sulfate soils (%)
Very high risk	710 $\pm$ 1600	32 $\pm$ 47	82 $\pm$ 17	670 $\pm$ 68	1200 $\pm$ 1400	98 $\pm$ 15
$\geq 4$	(0.90)	(0.92)	(0.79)	(0.82)	(0.80)	(0.77)
High risk	210 $\pm$ 750	5.4 $\pm$ 23	74 $\pm$ 18	700 $\pm$ 100	1200 $\pm$ 2400	92 $\pm$ 28
$\geq 3$	(0.95)	(0.98)	(0.76)	(0.78)	(0.80)	(0.73)
Mod. risk	170 $\pm$ 680	33 $\pm$ 18	54 $\pm$ 30	670 $\pm$ 100	790 $\pm$ 1800	24 $\pm$ 43
$\geq 2$	(0.93)	(0.99)	(0.75)	(0.69)	(0.74)	(0.90)
Low risk	35 $\pm$ 150	0.27 $\pm$ 5.2	37 $\pm$ 18	610 $\pm$ 180	122 $\pm$ 490	2.7 $\pm$ 16
$\geq 1$	(0.97)	(0.99)	(0.72)	(0.53)	(0.80)	(0.98)
Very low risk	2.5 $\pm$ 12	0	30 $\pm$ 7.0	160 $\pm$ 270	2.1 $\pm$ 15	0
$< 1$	(0.99)	(N/A)	(0.55)	(0.51)	(0.94)	(N/A)

value comparing the model with the variable removed to the actual model results. All input data and model outputs are publicly available (McKenzie et al. 2024).

Areas with the highest metal contamination risk from SGD (covering 4% of the global coast) are those with very high population density, high SGD, and the presence of acid sulfate soils, particularly around densely populated tropical regions (Fig. 3). From this, we identified five risk categories, where high population density, high presence of coastal mines and acid sulfate soils, very high percentage of untreated wastewater, and high SGD and sea-level rise characterized the highest risk group (Table 3). The three highest risk categories (moderate to very high) represent 36% of the global coastline and are particularly concentrated in the tropics. Climate change is projected to especially impact some of the highest risk regions (Calvin et al. 2023), further exacerbating conditions that promote metal mobilization and declining coastal water quality. For all risk categories, anthropogenic risk (AnthroRisk) and hydrogeological risk (HydroRisk) contributed between 31–45% and 55–69% to the total cumulative risk (CumRisk), respectively. The risk model has several sources of uncertainty, including the model variables selected and inherent uncertainties within each dataset. Furthermore, the model evaluates cumulative relative risk in a global context, meaning lower risk regions may still present risk from metal contamination. According to the sensitivity analysis, wastewater treatment, sea-level rise, and SGD had the greatest uncertainties associated with the final cumulative risk, whereas population density and presence/absence of mines had the least (Table 3).

While we know human actions (climate change, anthropogenic inputs) pose a metal contamination threat to the coastal aquifer and ocean, there are still remaining uncertainties especially regarding the magnitude and rate of local to global-scale impacts. Our risk analysis identified areas of particular interest

for future research such as the tropics and Indo-Pacific regions, as well as the Southeast coast of the United States. These regions should be considered when looking to quantify and understand the role of rapid change and multiple stressors on metal chemistry in SGD. Moreover, the confluence of local-scale environmental processes and metal sources are likely more complicated and thus highlight the importance for more field-based studies, especially for higher-risk locations.

### Conclusions and future directions

Anthropogenic pressure from climate change and contamination sources are driving dynamic changes to metal inputs and speciation within the STE and coastal ocean. STEs are an important interface along the land-ocean continuum, often acting as a filter for metals. However, climate change hazards (sea-level rise, increasing temperatures, precipitation changes, storm events, ocean acidification) are rapidly changing STE conditions and may impact metal speciation. Growing populations and development along coastlines put further pressure on coastal systems, both by modifying environmental conditions as well as increasing metal sources. These compounding effects will drive modifications in STE biogeochemistry (e.g., temperature, salinity, pH, oxygen concentrations, DOM, microbial communities), impacting metal speciation, behavior, and bioavailability.

SGD water and metal fluxes are understudied in many regions globally, particularly the Global South. Future research should aim to better understand metal behavior in the STE, influences on metal speciation (particularly adsorption and colloidal behaviors), and interactions between metal and other elemental biogeochemical cycles. In addition, more research is needed to resolve the role that fresh and saline SGD fractions play in metal delivery to the coastal ocean, as



well as improving our understanding of local to global scale SGD-driven metal fluxes.

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