

Review

A global-level assessment of gulls (*Larus* spp.) as bioindicators of trace elements in coastal ecosystems

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ABSTRACT. Anthropogenic activities and the demand for trace elements have risen, causing an increase in their environmental levels, which could affect biota. High levels of trace elements in living beings have been associated with toxicity, metabolic disorders, oxidative stress, and cancer. Seabirds, such as gulls, have been used as bioindicators of environmental pollution caused by anthropogenic sources. Gulls are widely distributed worldwide, usually occupying high trophic levels, and living close to humans. Among gulls, *Larus* spp. are omnivorous, predominantly carnivorous, nest on the ground, and capture live food or steal it opportunistically. The present work summarizes the data of trace elements found in eggs, feathers, blood, and feces, among other internal tissues, of several gull species of the genus *Larus* around the world. Most of the reports are from the Northern Hemisphere (70%), particularly from Europe and North America. The elements Cd, Hg, Mn, Pb, and Se are the most studied (54%), among which Hg represents about 19%. On the other hand, there is no information for lesser-known trace elements such as Rb, Ti, new technology elements (e.g. Ge, Re, Ta), rare earth elements (e.g. Ce, La, Y), or elements of the platinum group (e.g. Os, Ir, Ru). Even though *Larus* spp. is a suitable bioindicator of chemical contamination in marine ecosystems, only 28 of the 53 species of the *Larus* genus have been used on trace elements pollution. Future research should address lesser-known elements which are increasingly used by new technologies.

Keywords: *Larus*; seabirds; gull; trace elements; bioindicators; marine pollution

INTRODUCTION

Human life depends on oceans for survival, but anthropogenic activities have altered the health of most oceanic ecosystems (Halpern et al. 2008). Ocean pollu-

tion, overfishing, species introduction, diseases, population growth, and climate change have been major factors affecting coastal ecosystems (Boesch 2001). Chemical pollution produced by humans is hard to handle because 95% of all manufactured goods come

from a great diversity of chemistry industries, such as agrochemicals, metallurgy, mining, electronics, and pharmaceuticals, among others (Wang et al. 2020). Most of the pollutants reach aquatic ecosystems, becoming a problem of global concern as they increase along with population and anthropogenic activities. Due to the toxicity of trace elements and their deleterious effects on fauna (e.g. damage to the internal organs and metabolic disorders), their effects on wildlife are of great concern among researchers (Takekawa et al. 2002). That is why studies of heavy metals in wildlife are important, adding information on man-made environmental changes and their impact on ecosystems (Zhou et al. 2001, Celis et al. 2014, Espejo et al. 2018a).

Coastal pollution can be studied through the chemical compound's bioaccumulation in biota, which are used as bioindicators to screen the health of ecosystems (Parmar et al. 2016). Shorebirds, such as gulls, are an important fauna of coastal ecosystems (Muñoz-Cifuentes et al. 2003) because they are high-trophic level species that may accumulate persistent chemical elements, and sensitive organisms can express their effects in a short time (Lewis et al. 1993). Moreover, coastal birds have a wide distribution (De Moreno et al. 1997) and live long enough (ICES 2003), which makes them suitable species to be used as bioindicators of local pollution environments (Lewis & Furness 1991, Muñoz-Cifuentes et al. 2003).

The Laridae family is a large group of gull species worldwide, with important morphological and ecological differences (Del Hoyo et al. 1996). Morphologically gulls are uniform in shape, with heavy bodies, long wings, medium-length necks, moderately long tarsi, fully webbed feet, and usually of a white body with a dark cloak, which varies from silver gray to black (Baker et al. 2007). Generally, the tail is rounded in all species, with long, moderately large, and slightly rounded wings that help them to have a lightweight flight with great maneuverability. In almost all aspects, *Larus* spp. are generalists, i.e. birds less specialized about foraging methods or type of food, making cannibalism a common practice among large gulls (Del Hoyo et al. 1996). For that reason, they are easily adapted to occupy a wide variety of ecosystems, ranging from polar to desert. Gull weight varies from 0.1 kg to more than 2 kg, and although gulls do not exhibit sexual dimorphism in plumage, females are slightly smaller than males. Larger gull species live in colder zones, and the smaller ones in more temperate or tropical regions. They also have salt glands, allowing them to adapt to marine habitats (Del Hoyo et al. 1996).

Some *Larus* spp. have become a pest in certain places around the planet, causing problems around landfills, airports, agricultural areas, and communities, where their populations have increased dramatically (Sinclair et al. 2002). However, *Larus* spp. can be very useful as bioindicators of environmental problems because these birds can become an early warning of pollution problems and be a tool to evaluate the health of ecosystems (Del Hoyo et al. 1996). Table 1 lists the species of *Larus* spp., their habits, and their diet preferences.

Trace elements can be divided into non-essentials and essentials. Non-essential trace elements such as mercury (Hg), cadmium (Cd), arsenic (As), or lead (Pb) do not participate in the animal metabolism at all, being toxic when its levels increase in the organism, causing acute toxicity that affects the central nervous system, kidney, and liver (Castillo et al. 2005). Essential trace elements, copper (Cu) or zinc (Zn), are elements required for individual metabolism in small concentrations, remaining vital in the physiological and biochemical functions of living beings (Tahri et al. 2005). However, all metals can be powerful toxic, and even carcinogens, depending on the levels at which they have presented and the chemical nature of the metal (Tchounwou et al. 2012). Population growth and industrial, agricultural, mining, and transport increase are the main sources of trace elements (Ouyang et al. 2006). Due to these elements' accumulative nature and persistence, they can be found in the water, air, and soil, usually detected in flora and fauna (Zhao et al. 2006). The effect of well-known trace elements (e.g. Pb, Cd, Hg) is more severe in species at the top of food chains as they tend to bioaccumulate pollutants, such as gulls and humans (Burger et al. 2000).

Studies conducted in recent decades on analyzing metal concentrations in gulls in coastal ecosystems worldwide are fragmented and insufficient (Otero-Pérez 1998, Ninomiya et al. 2004, Agusa et al. 2005, Grajewska et al. 2019, De Medeiros-Costa et al. 2021). A systematic review of the existing literature about the levels of trace elements in different biotic matrices of *Larus* spp. was conducted. Thus guano, feathers, eggs, blood, stomach contents, and internal organs were considered. We used Direct, Springer, Scopus, and Web of Science databases. Different keywords were used: "trace element", "heavy metal", "trace metals", "mercury", "aluminum", "arsenic", "cadmium", "lead", "zinc", "copper", "pollution", "persistent pollutants", "monitoring", "biomonitoring", "gull", "seabirds", "eggs", "blood", "guano", "droppings", "feathers", "tissues", and "organs". Also, the list of references for

each publication was reviewed to identify additional documents on the issue not previously found. Selection criteria were for trace element levels based on dry weight (dw) based on studies performed *in situ*. Trace element burden on the eyeball, gallbladder, intestine, pancreas, skin, stomach, subcutaneous fat, uropygial gland, claws, feces, and stomach content, were not considered here because they were reported only by a single study and based on wet weight.

Exposure to trace elements

Trace element concentrations measured in different biotic matrices of different species of gulls are indicated in Table S1 (see Supplementary material). Most trace element contents in *Larus* spp. were reported from Northern Hemisphere compared to Southern Hemisphere (Fig. 1). While 16 reports are from Northern Hemisphere, particularly from USA and Europe. In contrast, only seven are from Southern Hemisphere, the majority from Argentina, Brazil, and Chile.

The most common matrices reported were eggs (n = 216), feathers (n = 87), liver (n = 54), kidney (n = 35), and blood (n = 20). Eggs have two benefits; they are non-lethal samples and are easily collected. When breeding occurs, females ingest large amounts of food and mobilize proteins and fat to form eggs and the pollutants attached to them (Walker 1994, Becker et al. 2001). In addition, bird eggs have been used as bioindicators of long-term studies at temporal and spatial scales (Braune et al. 2002). In addition, feathers are commonly used as non-lethal biotic matrices. The trace elements ingested through a bird's diet are accumulated in the feathers through their transport by blood (Arcos et al. 2002). However, the mechanisms involved in transporting the metal still need to be better understood.

The burden of essential trace elements

Copper (Cu): this metal has been measured in 12 species from 15 countries and territories (Table S1). The lowest concentration was found in *L. genei* eggs ($0.33 \mu\text{g g}^{-1}$, Hernández et al. 1999), while the highest Cu levels were found in *L. cachinnans* feces ($60.1 \mu\text{g g}^{-1}$), both from Spain (Otero-Pérez 1998), followed by *L. michahellis* eggshells ($54.4 \mu\text{g g}^{-1}$) from Turkey (Durmaz et al. 2016). The liver was the most sampled tissue studied, followed by feathers and the kidney. Only based on liver tissue, *L. ridibundus* from Poland and Spain showed the highest Cu contents (Hernández et al. 1999, Gómez et al. 2004, Orłowski et al. 2007), while the lowest were found in *L. crassirostris* (Kim &

Oh 2014a) from South Korea and *L. dominicanus* from New Zealand (Numata et al. 2008). Feather is the most common non-lethal tissue to monitor Cu concentrations. This metal accumulates mostly in the liver, kidney, and eggshell. In contrast, eggs, bones, and blood seem to be tissues less indicated to bioaccumulate Cu (Fig. 2a). The contents of Cu found in gull feathers are higher in Chile, South Korea, Brazil, and Portugal. Cu pollution occurs due to natural and anthropogenic sources, including Cu factories, Cu mining, fossil fuels combustion and wastes, domestic wastewater, phosphate fertilizer production, wood production, volcanoes, windblown dust, and others (Rehman et al. 2019).

Zinc (Zn): this metal has been measured in 13 species from the same number of countries. The lowest Zn levels ($0.29 \mu\text{g g}^{-1}$) have been found in *L. saundersi* eggshells from China (Yang et al. 2020). On the other hand, the highest Zn contents were found in *L. michahellis* eggshells ($427 \mu\text{g g}^{-1}$) from Turkey (Durmaz et al. 2016), followed by Spain *L. cachinnans* feces ($305.1 \mu\text{g g}^{-1}$) and Portugal *L. michahellis* feathers ($260.1 \mu\text{g g}^{-1}$). The liver was the most studied organ to measure Zn concentration, followed by feathers, muscles, and kidneys. Data show that the levels of Zn are higher in the kidneys and liver of gulls. It is well known that Zn pollution is mainly associated with mining-related activities and metallurgical industries over an extended period (Yin et al. 2015).

Iron (Fe): this metal has been evaluated in six *Larus* spp. from the same number of studies from countries in North America, Asia, and Europe. The lowest Fe levels ($2.09 \mu\text{g g}^{-1}$) are found in *L. crassirostris* stomach content from South Korea (Kim & Oh 2014b), whereas the highest Fe levels ($4,056 \mu\text{g g}^{-1}$) were reported in feathers of the same species and location (Kim et al. 2013). Feathers were the most used biotic matrix to study this element; however, it seems that the liver is a tissue in which metal tends to accumulate the mostly in the liver. Fe is an essential nutrient of most organisms, but high Fe levels in tissues have been associated with cancer, liver and heart disease, diabetes, hormonal abnormalities, and immune system dysfunctions (Fraga & Oteiza 2002).

Manganese (Mn): this metal has been measured in 14 *Larus* spp. from 12 countries, including the Southern Hemisphere (Brazil, South Africa, and Namibia of the Southern Hemisphere). The lowest Mn levels ($0.00124 \mu\text{g g}^{-1}$) were reported in *L. pipixcan* kidneys from Minnesota, USA (Burger 1993), while the highest Mn levels are found in *L. crassiro* feathers from South Korea ($122 \mu\text{g g}^{-1}$; Kim et al. 2013) and *L. michahellis*

Table 1. List of species of *Larus* spp. (common and scientific names), habitat and diet preferences.

Species	Habitat	Diet
Herring gull (<i>L. argentatus</i>)	Very abundant on European coasts but more frequently found on the Atlantic Ocean and Mediterranean Sea shores (Del Hoyo et al. 1996).	This species is opportunistic, mainly feeding fish and garbage (Del Hoyo et al. 1996).
Armenian gull (<i>L. armenicus</i>)	This species breeds from Armenia to western Turkey and northwest Iran, wintering south to the eastern Mediterranean, northern Red Sea, and northern Persian Gulf (IUCN 2022).	Its poorly known diet may consist primarily of fish and terrestrial invertebrates, amphibians, reptiles, and rodents (Del Hoyo et al. 1996).
Laughing gull (<i>L. atricilla</i>)	Strictly on the American coast, it breeds throughout North Carolina (USA), Mexico, Venezuela, and even the Caribbean.	Invertebrates, aquatic insects, fish, waste, and garbage (Del Hoyo et al. 1996).
Audouin's Gull (<i>L. audouinii</i>)	Endemic to the Mediterranean. The largest colonies are in the Delta of the Ebro and Chafarinas Islands, Spain.	Mainly piscivorous. In addition, these birds can take advantage of the freshwater resources of the rice paddies of the Ebro Delta (Navarro et al. 2010).
Caspian's gull (<i>L. cachinnans</i>)	It usually lives on the Black Sea and the Caspian Sea shores, extending from Central Asia to the northwest of China. Their populations have expanded to the north and west of Europe, mainly in Poland and eastern Germany. Some have migrated to the Red Sea and the Persian Gulf, Norway, and Denmark (Hume 2007).	Scavengers and predators, with a very varied diet. During the breeding season, they often eat rodents, such as ground squirrels (Olsen & Larsson 2004).
California gull (<i>L. californicus</i>)	It inhabits the west coast of North America, from Canada to Mexico.	It feeds on insect larvae, eggs, young birds, rodents, and garbage (Del Hoyo et al. 1996).
Common gull (<i>L. canus</i>)	This species presents three subspecies, which are distributed across Europe and Asia.	These birds are omnivores, but they hunt small living prey and even can scavenge (Olsen & Larsson 2004).
Black-tailed gull (<i>L. crassirostris</i>)	It is originally from East Asia, including China, Taiwan, Japan, and Korea. Occasionally, this species can be found in Alaska and northeastern North America.	It feeds mainly on small fish, mollusks, crustaceans, and scavengers. It often pursues commercial fishing vessels and steals food from other seabirds (Del Hoyo et al. 1996).
Ring-billed gull (<i>L. delawarensis</i>)	This species inhabits North America and Central America, where it can be found on beaches, rivers, estuaries, marshes, and landfills.	It is a very opportunistic species, with a wide diet including fishes, insects, earthworms, crabs, eggs, rodents, birds, and organic waste (Del Hoyo et al. 1996).
Kelp gull (<i>L. dominicanus</i>)	Not a migratory, it is present widely in the Southern Hemisphere, including Antarctica, Australia, New Zealand, South Africa, and the southern South American continent (Del Hoyo et al. 1996).	Mainly piscivorous, although it also feeds on boat waste and landfills (Del Hoyo et al. 1996).
Heuglin's gull (<i>L. fuscus heuglini</i>)	This species inhabits northern Russia to Finland but also migrates to Southwest Asia and East Africa during winter.	It feeds mainly on mollusks, worms, and crustaceans (Olsen & Larsson 2004).
Slender-billed gull (<i>L. genei</i>)	Inhabits mainly the Mediterranean, the Black Sea, and the Western Indian Ocean. Most populations are migratory, moving in winter to the north of Africa and a few toward Western Europe (Del Hoyo et al. 1996).	Mainly fish (c. 50% of the diet), insects, and marine invertebrates (Del Hoyo et al. 1996).
Glaucous-winged gull (<i>L. glaucescens</i>)	Lives on the shores of the North Pacific, being very common in the coastal towns and villages.	It is characterized by a varied diet, including live animals, dead animals, and organic waste (Olsen & Larsson 2004).
Hartlaub gull (<i>L. hartlaubii</i>)	Endemic to the western coast of South Africa and Namibia (Teele 1992).	It is omnivorous, feeding on debris and small prey (Teele 1992).
Heermann's gull (<i>L. heermanni</i>)	Commonly found in the USA, Mexico, and extreme southwestern British Columbia, but it usually nests on Isla Rasa, Gulf of California (IUCN 2020).	It feeds on small fishes, marine invertebrates, lizards, insects, waste, and carrion (IUCN 2020).
Heuglin's gull (<i>L. heuglini</i>)	It lives in the tundra of northern Russia, from the Kola Peninsula to the east of the Taymyr Peninsula. These birds migrate south to spend the winter (in southwest Asia and east Africa).	They feed primarily on mollusks, worms, and crustaceans (Olsen & Larsson 2004).

Continuation

Species	Habitat	Diet
Glaucous gull (<i>L. hyperboreus</i>)	This species is the most common bird in the Arctic (Anker-Nilssen et al. 2001).	It plays an important role as a top predator in the European Arctic (Anker-Nilssen et al. 2001).
Great black-backed gull (<i>L. marinus</i>)	On coastal areas in the northwest of Russia, shores of Scandinavia, the Baltic Sea, the north coast of France, the United Kingdom, Ireland, in the northern part of the Atlantic, south of Iceland, Greenland, and on the Atlantic coasts of Canada and the USA (Del Hoyo et al. 1996).	It is an opportunistic species, feeding on human waste (more than half of its diet). Also, it captures fish near the water's surface (Del Hoyo et al. 1996).
Mediterranean gull (<i>L. melanocephalus</i>)	It inhabits almost the entire region of Europe, even found on the Black Sea coast of Ukraine.	It feeds on insects, fishes, and rodents; during the breeding season, it can feed on mollusks, insects, worms, seeds, and organic waste (Del Hoyo et al. 1996).
Yellow-legged gull (<i>L. michahellis</i>)	It is a large gull in Europe, the Middle East, and North Africa.	It is omnivorous and opportunistic foragers (Zorrozueta et al. 2020).
Western gull (<i>L. occidentalis</i>)	The west coast of North America.	At sea, it feeds on fish and invertebrates. On land, it feeds on seal and sea lion carcasses and road kill, among others (Emslie & Messenger 1991).
Black-headed gull (<i>L. ridibundus</i>)	It experienced an increase in its population, mainly due to the colonization of the artificial wetlands created in European countries (Cramp 1998).	This bird's diet is relatively wide, including food on land under cultivation, municipal landfills, and different aquatic organisms (Cramp 1998).
Sabine's gull (<i>L. sabini</i>)	It breeds in the Arctic through northernmost North America and Eurasia. It migrates south and winters at sea off western South America in the cold waters of the Humboldt Current. At the same time, Greenland and eastern Canadian birds cross the Atlantic through the westernmost fringes of Europe to winter off southwest Africa in the cold waters of the Benguela Current (Del Hoyo et al. 1996).	Small fish, birds, macroinvertebrates, eggs, and carrion (Olsen & Larsson 2004).
Saunders's gull (<i>L. saundersi</i>)	This species breeds in eastern China and the west coast of Korea (IUCN 2022).	They catch mudskippers, crabs, fish, and worms and steal food from other bird species (IUCN 2022).
Thayer's gull (<i>L. thayeri</i>)	It is native to North America, breeding in the Arctic islands of Canada during winter and on the Pacific coast, ranging from southern Alaska to the Gulf of California (Olsen & Larsson 2004).	Omnivores: eat small fish, crustaceans, mollusks, carrion, eggs, young birds, and garbage (Del Hoyo et al. 1996).
Brown-hooded gull (<i>L. maculipennis</i>)	It mainly inhabits South America, specifically on the coast of Chile, Argentina, Uruguay, and the Falkland Islands (Del Hoyo et al. 1996).	It feeds on insects, although those inhabiting Chile's shores feed mainly on fish (Del Hoyo et al. 1996).
Franklin's gull (<i>L. pipixcan</i>)	This species migrates long distances from North America, where it is reproduced in the marshes of the prairies relatively unchanged, up to the coasts of South America (Burger & Gochfeld 2009).	It feeds on small fish, marine invertebrates, earthworms, and insects during the breeding season (Burger & Gochfeld 2009).

eggshells from Turkey (73.47 $\mu\text{g g}^{-1}$; Durmaz et al. 2016). Nevertheless, comparing the Mn tissue distribution along the studies, it seems this metal accumulates the most in the liver and kidneys. Again, feathers are the most common tissue used to monitor this element, followed by eggs and the liver. Excess dietary exposure to Mn in humans can cause deleterious effects on the central nervous system leading to a progressive disorder in the population (e.g. Parkinson's disease) (Kwakye et al. 2015).

Nickel (Ni): this metal is usually found at very low levels in the environment, but its vast industrial usage has caused it to spread worldwide. Reports about Ni contents in *Larus* spp. have shown the following decreasing order: Turkey (30.72 $\mu\text{g g}^{-1}$ in *L. michahellis* eggshells; Durmaz et al. 2016) > Brazil (5.92 $\mu\text{g g}^{-1}$ in *L. dominicanus* feathers; Barbieri et al. 2010) > Portugal (3.31 $\mu\text{g g}^{-1}$ in *L. audouinii* feathers; Laranjeiro et al. 2020) > China (3.19 $\mu\text{g g}^{-1}$ in *L. saundersi* eggshells; Yang et al. 2020) > Iran (2.27 $\mu\text{g g}^{-1}$ in *L. heuglini* livers; Mansouri et al. 2012) > South

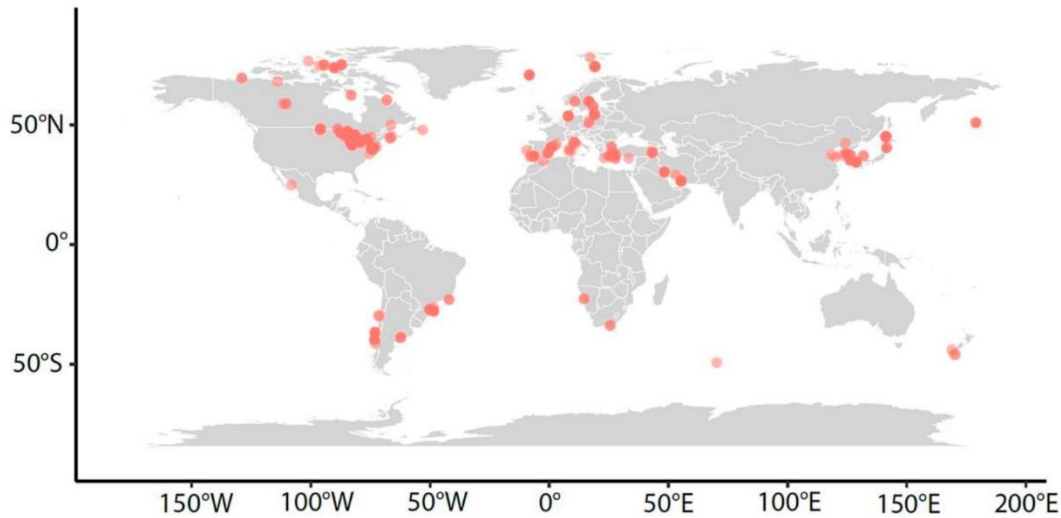


Figure 1. Geographical distribution of the studies performed on *Larus* spp. around the world.

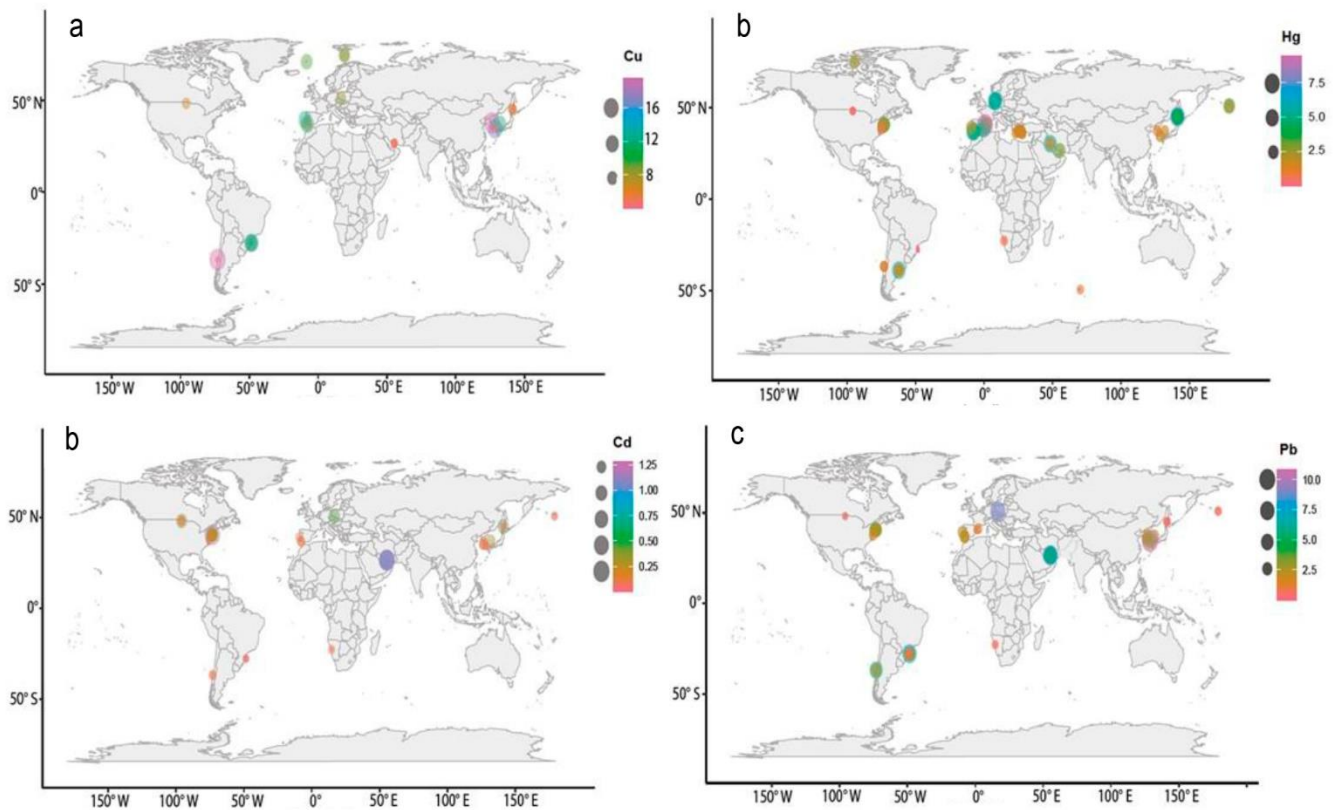


Figure 2. Map of the most studied metals in feathers of *Larus* spp.: a) copper; b) mercury; c) cadmium; d) lead.

Africa ($1.4 \mu\text{g g}^{-1}$ in *L. dominicanus* eggshells; Van Aswegen et al. 2019). Feathers, followed by eggshells, are the most common sample tissues studied. Even though Ni is an essential metal for animals, excessive Ni levels tend to change biological structures and systems, causing leading to cancer, liver and heart

disease, diabetes, and other anomalies (Rehman et al. 2018).

Molybdenum (Mo): there is very scarce information on Mo burden in *Larus* spp. worldwide. It is an essential element that participates in the biological process of nitrogen fixation in organisms, presenting relatively

low toxicity to humans (Barceloux 1999). Studies from Brazil, South Africa, and Japan have reported the Mo burden in the liver (Henrique-Pedrobom et al. 2021), eggs (Van Aswegen et al. 2019), and several other tissues (Agusa et al. 2005), respectively. The lowest Mo levels ($0.025 \mu\text{g g}^{-1}$) have been found in *L. crassirostris* eggshells from Japan (Agusa et al. 2005). In contrast, kidneys and liver of the same species and location exhibited higher Mo concentrations (3.81 and $2.45 \mu\text{g g}^{-1}$, respectively).

Chromium (Cr): this metal has been measured in 15 *Larus* spp. from 11 countries worldwide. The lowest Cr concentrations were found in *L. argentatus* eggs from New York (Burger & Elbin 2015) and *L. hartlaubii* feathers at Swakopmund, Namibia (Burger & Gochfeld 2001). Eggs of *L. dominicanus* from Swartkops (South Africa) exhibited the highest Cr contents ($18 \mu\text{g g}^{-1}$) (Van Aswegen et al. 2019), much higher than the global average ($1.12 \pm 2.02 \mu\text{g g}^{-1}$). This metal exhibits a different pattern than most metals, but Cr accumulates mainly on eggshells, blood, and feathers. Feathers and eggs are the most studied tissues, but Cr accumulates mainly in eggshells, followed by blood and feathers. Bioaccumulation of Cr in organisms can produce mutagenesis and carcinogenesis at certain levels (Sharma et al. 2021).

Magnesium (Mg): even though magnesium (Mg) is not a trace element, it is the fourth most abundant element having an important role in animals (Jahnen-Dechent & Ketteler 2012). Two studies have only reported this element in *L. armenicus* from Turkey (Durmuş et al. 2018) and in feathers and eggs of *L. pipixcan* from the USA (Custer et al. 2007), with Mg concentration in feathers higher in Turkey than in the USA. Due to the low number of studies carried out in *Larus* spp. so far and the tissues sampled, comparing the burden of tissues was not possible this time.

The burden of non-essential trace elements

Mercury (Hg): this metal has been the most studied in *Larus* spp. with 22 countries and territories where Hg has been monitored in 26 species (Table S1). The lowest Hg concentrations have been reported in *L. dominicanus* feathers from Brazil (Ebert et al. 2020) and *L. dominicanus* eggshells from South Africa (Van Aswegen et al. 2019). On the other hand, the highest Hg concentrations in gull feathers have been found in coastal areas of the northern Mediterranean Sea (Fig. 2b). Some areas of Spain, Portugal, Japan, Germany, and Argentina have reported Hg concentrations above $5 \mu\text{g g}^{-1}$, higher than the world average ($0.47 \mu\text{g g}^{-1}$). Research from the Arctic and Canada have found Hg of

4.9 and $1.56 \mu\text{g g}^{-1}$ in eggs of *L. hyperboreus* (Braune et al. 2016) and *L. argentatus* (Hebert et al. 2021), respectively. The tissue distribution indicates that Hg bioaccumulates the most in the liver and feathers. Hg is one of the most toxic elements with global distribution due to centuries of emissions and redistribution. This metal can form salts in two ionic states, mercury (I) and mercury (II); the second is much more common in the environment (Boening 2000). However, the organic mercury form (mostly methylmercury, MeHg (CH_3Hg^+)) bioaccumulates and biomagnifies through the food web (Clayden et al. 2017). MeHg is also detrimental to humans, associated with nervous system damage in adults, impaired neurological development in infants and children, and endocrine disruption in wildlife (Bergman et al. 2012, Rice et al. 2014).

Cadmium (Cd): this metal is very toxic to biota and is produced by primary metal industries, batteries, foods, soil, and cigarette smoke, among others (Saini & Dhania 2020). It has been linked to bone disease, renal damage, and several forms of cancer. Sixteen species from the same number of locations have been tested for Cd in feathers, eggs, liver, and kidneys. The lowest Cd levels ($0.0003 \mu\text{g g}^{-1}$) are reported in eggs of *L. argentatus* from New York, USA (Burger & Elbin 2015). The Arctic is where the highest Cd concentrations ($99.6 \mu\text{g g}^{-1}$) have been reported in *L. hyperboreus* (Braune & Scheuhammer 2008, Malinga et al. 2010) and *L. crassirostris* kidneys from Japan ($14 \mu\text{g g}^{-1}$, Agusa et al. 2005), with some other hotspots found in *L. dominicanus* liver from Chile ($9.84 \mu\text{g g}^{-1}$, Cortés & Luna-Jorquera 2011) and *L. ridibundus* liver from Spain ($1.35 \mu\text{g g}^{-1}$, Gómez et al. 2004). The highest Cd contents in gull feathers are from areas of Iran and the USA (Fig. 2c). The reports from 10 different gull tissues studied worldwide show that Cd accumulates mainly in the kidneys and liver.

Lead (Pb): this metal increased its presence in the environment because it was profusely used as an additive in gasoline during the past decades. During the '90s, some regulations banned the use of Pb in gasoline, and thus Pb emissions have been reduced worldwide (Boutron et al. 1995). The lowest concentration is in the eggshells of *L. dominicanus* from South Africa ($0.00006 \mu\text{g g}^{-1}$, Van Aswegen et al. 2019), eggs, and feathers of *L. argentatus* from a strongly USA urbanized area (Burger 1993). On the other hand, the highest Pb concentrations are found in gull bones ($42.32 \mu\text{g g}^{-1}$), brain, and kidneys of *L. ridibundus* from Poland (Orłowski et al. 2007), followed by the feather of *L. crassirostris* from South Korea (Kim et al. 2013), and *L. heuglini* kidneys from Iran (Hoshiyari et al.

2012). Using gull feathers as non-lethal tissues to study Pb pollution, the evidence indicates that South Korea, Poland, Iran, Brazil, and Chile have higher Pb contents in *Larus* spp. (Fig. 2d). Nevertheless, there are some peaks of Pb levels in the bones and brain of gulls, although kidneys and feathers seem to accumulate Pb mostly.

Arsenic (As): this metalloid is a well-known carcinogen and one of drinking water's most significant chemical contaminants worldwide (WHO 2003). This element has been measured in 11 *Larus* spp. from 10 different regions worldwide. Even as the average concentration in the Earth's crust is about $5 \mu\text{g g}^{-1}$, its concentration has increased due to anthropogenic activities such as mining or mining-related operations. The lowest As levels ($0.0002 \mu\text{g g}^{-1}$) are found in eggs of *L. argentatus* from New York, USA (Burger & Elbin 2015), whereas Brazil ($5.9 \mu\text{g g}^{-1}$, Henrique-Pedrobom et al. 2021), USA (Burger & Gochfeld 1997) and Spain (Hernández et al. 1999) have reported the highest As contents in *L. dominicanus*, *L. argentatus*, and *L. audouinii*, respectively. The most common tissues used are eggs, feathers, and the liver. The liver and blood seem to be the tissues with major As bioaccumulation.

Aluminum (Al): there are few investigations on Al concentration in gulls. Al is a serious environmental toxicant, and it is detrimental to biota. Al has been measured in five *Larus* spp. from Brazil, the USA, South Korea, and Portugal (Table S1). Feathers are the most studied tissue, with the highest concentration reported. The lowest Al levels were found in the blood of *L. crassirostris* from South Korea ($1.34 \mu\text{g g}^{-1}$; Kim et al. 2013). The highest Al levels ($3,547.56 \mu\text{g g}^{-1}$) were found in feathers of *L. crassirostris* from South Korea (Kim et al. 2013), followed by $192.7 \mu\text{g g}^{-1}$ in *L. audouinii* from Portugal (Laranjeiro et al. 2020).

Barium (Ba): in animals, Ba chronic exposure leads to cardiovascular and kidney diseases and metabolic and neurological disorders (Kravchenko et al. 2014). However, information about Ba bioaccumulation and its effects on gulls is scarce. Only a few countries (Turkey, Japan, USA, and Brazil) have reported Ba concentration in *Larus* spp. The lowest Ba levels were found in the liver of young chicks (*L. crassirostris*) from Japan ($0.017 \mu\text{g g}^{-1}$; Agusa et al. 2005), while the highest concentration was found in *L. michahellis* eggshells from Turkey ($>700 \mu\text{g g}^{-1}$; Durmaz et al. 2016).

Antimony (Sb): this is a metal of concern because it can potentially be a carcinogenic agent, although there is a huge gap in information on Sb bioaccumulation and bioavailability (Obiakor et al. 2018). There are only

some studies from Japan in *L. crassirostris* (Agusa et al. 2005), from South Africa in *L. dominicanus* (Van Aswegen et al. 2019), and Turkey in *L. michahellis* (Durmaz et al. 2016). The results indicated that the order of levels of Sb in eggshells of *Larus* spp. is Turkey ($0.092 \mu\text{g g}^{-1}$) > Japan ($0.03 \mu\text{g g}^{-1}$) > South Africa ($0.02 \mu\text{g g}^{-1}$).

Strontium (Sr): this metal has been associated with bone cancer in humans, while in animals has showed to induce some alterations in bone mineralization and renal functions (Cohen-Solal 2002). Different tissues have been studied worldwide (Brazil, Japan, the USA, and South Africa) to evaluate Sr levels (*L. dominicanus*, *L. crassirostris*, and *L. pipixcan*, respectively). The levels of Sr in gull eggshells and bones showed an order of magnitude higher than any other tissues.

Silver (Ag): this metal has been used widely as an antimicrobial. Ag levels have been assessed in *L. dominicanus* from South Africa ($0.5 \mu\text{g g}^{-1}$, eggshells; $0.28 \mu\text{g g}^{-1}$ eggs; Van Aswegen et al. 2019) and in *L. crassirostris* from Japan ($0.005 \mu\text{g g}^{-1}$ eggshells, $0.008 \mu\text{g g}^{-1}$ eggs; Agusa et al. 2005). In wildlife, there is no evidence showing Ag biomagnification from marine or freshwater ecosystems, although certain toxicity at low levels has been linked to Ag (Ratte 1999).

Tin (Sn): the highest Sn contents have been reported in eggs of *L. pipixcan* from Minnesota ($2.1 \mu\text{g g}^{-1}$, Custer et al. 2007) and liver of *L. argentatus* ($1.366 \mu\text{g g}^{-1}$) and *L. marinus* ($0.838 \mu\text{g g}^{-1}$) near the John F. Kennedy International Airport, USA (Burger et al. 2000). The concentrations reported at the other two studies have shown lower concentration at Turkey (*L. armenicus* feathers, $0.012 \mu\text{g g}^{-1}$) and Namibia (feathers of *L. dominicanus* and *L. hartlaubii*). The most common tissues studied are feathers.

Vanadium (V): only a few studies have reported this metal in three gull species. The lowest V levels are reported in *L. dominicanus* liver ($0.2 \mu\text{g g}^{-1}$) from Brazil and several tissues of *L. crassirostris* from Japan (all concentrations $<0.13 \mu\text{g g}^{-1}$). The highest V levels have been found in *L. dominicanus* eggshells ($170 \mu\text{g g}^{-1}$) from South Africa (Van Aswegen et al. 2019) and *L. michahellis* eggshells ($4.91 \mu\text{g g}^{-1}$) from Turkey (Durmaz et al. 2016). Data showed no clear tendency about which gull tissue concentrates this metal.

The burden of less-known trace elements

There is no information on rubidium (Rb) and titanium (Ti) bioaccumulation in *Larus* spp. A few elements were reported by a single study (Table S1). For instance, cesium (Cs) was evaluated in *L. crassirostris*

from Japan (Agusa et al. 2005); in Turkey, boron (B) was assessed in *L. michahellis* (Durmaz et al. 2016); lithium (Li) in *L. americanus* (Durmuş et al. 2018). Only thallium (Tl) and uranium (U) were reported in *L. dominicanus* from South Africa (Van Aswegen et al. 2019).

Analytical methods used to quantify trace elements

Most studies have quantified trace element concentrations in biotic matrices using different methodologies, which vary according to their sensitivity and specificity for metals (Table S1). According to the studies on *Larus* spp. reviewed here, the analysis of trace elements by atomic absorption spectrophotometry (AAS) stands out with 48.7%, the graphite furnace atomic absorption spectroscopy (GFAAS) with 24.3%, the inductively coupled plasma-mass spectrometry (ICP-MS) with 19.8%, the atomic fluorescence spectrophotometry (AFS) with 6.4%, and the electrochemical analyzer equipped with a BAS cell with 0.6%. Particularly, Hg is 85.2%, determined by cold vapor atomic absorption spectrophotometry (CVAAS), and 14.7% by direct mercury analyzer (DMA) due to Hg being highly volatile.

SOME REMARKS

According to the data analyzed here, most studies on trace element contents in *Larus* spp. have usually focused on Hg, Cd, Pb, Mn, and Se (53.98% of the total studies). This review revealed that Hg is the most studied element (18.52%), whereas U is the less investigated.

Concerning the geographical distribution of the studies, most of them have been carried out in the Northern Hemisphere, particularly between 35 and 45°N, with few studies in the Southern Hemisphere. Sixteen regions of the Northern Hemisphere have reported data on trace elements in *Larus* spp. (Canada, USA, China, German, Greece, Iran, Italy, Japan, Mexico, Norway, Poland, Portugal, South Korea, Spain, Turkey, and the Arctic), while only seven regions of the Southern Hemisphere report some data from Argentina, Brazil, Chile, Namibia, New Zealand, South Africa, and Antarctica. Future studies should focus on trace element contents in gulls that inhabit the coasts of Central America, Africa, the Middle East, and Australia.

Of the 53 species that integrate the *Larus* genus (<https://www.gbif.org/>), only 28 species have been used as a bioindicator of trace elements pollution, being necessary to include studies in the rest of 52% of the species. Within these non-study species, it is important

to pay attention to those that are migratory, such as Franklin's gull, which migrates between Canada and southern Chile, because they might transport these pollutants from the Northern to Southern Hemisphere, perhaps acting as a global biovector of dispersion of chemical elements of anthropogenic origin.

Very few studies assess trace elements' biological effects on species of *Larus* spp. Of the 84 publications reporting trace element concentrations in gulls, only 12% have evaluated some biological effects. This lack of information reflects the real need for interdisciplinary teams in environmental sciences, a previously stated issue by Kaufmann & Cleveland (1995).

Despite other endangered species, *Larus* spp. can be used profusely to study the levels of metals in different internal organs and relate them to their biological effects on the organism. Most studies have focused on measuring the levels of exposure in different biotic matrices. The concentration of trace elements in tissues and organs of *Larus* spp. may have great toxicological importance. In humans, diseases related to metal pollution are quite known (Nordberg & Nordberg 2016). All species of *Larus* spp. are omnivorous, opportunistic foragers and are not protected under any conservation regulations, making them ideal for studying the health effects of trace elements, which could be extrapolated to humans. More studies with biomarkers are required to evaluate the risks associated with the levels of these contaminants. Further toxicokinetics studies of trace element levels in gulls, including other tissues and organs, are needed to understand the overall toxicity in seabirds better.

LIMITATIONS AND SUGGESTIONS

Rapid screening technology for detecting trace elements in biotic samples is desirable for researchers. It is crucial to implement uniform analytical protocols to help unify the data and make it more comparable. XRF is a fast and non-destructive method in simultaneously measuring the total concentration of multi-elements. Although XRF does not have sensitive detection limits of most trace elements as ICP-OES or ICP-MS, modern XRF analyzer is characterized by the limited preparation required for solid samples, non-destructive analysis, portability, high speed, lower waste production, low costs, and multi-elemental determination, which make it suitable for a rapid screening tool (Celis et al. 2022).

This review states that *Larus* spp. are suitable organisms to study trace elements since they are species distributed throughout all regions and environments of the planet, abundant, and not protected by conservation

laws like other species of seabirds. Most gulls are on the trophic chain's upper side and depend on numerous species for food. On the other hand, the human population will increase marine-derived protein consumption. Since gulls include fish in their diets, which humans consume, these seabirds might be used as a bioindicator for human health and exposure assessment. In addition, gulls have special adaptive characteristics, exhibit omnivorous feeding habits, and live close to humans; thus, any possible toxic effects caused by trace elements can be reasonably extrapolated to humans.

Future research should address lesser-known elements which are being increasingly used by new technologies, i.e. the platinum group (Pt, Pd, Rh, Os, Ir, and Ru), rare earth elements (La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Yb and Lu), including other elements which can be a threat to wildlife, such as tellurium (Te), germanium (Ge), gallium (Ga), Li, indium (In), niobium (Nb), tantalum (Ta) (Espejo et al. 2018a,b, Celis et al. 2020). The evaluation of trace elements mixing with other environmental contaminants should also be an issue to be explored. Much of the research has focused on reporting concentrations and environmental monitoring. However, few studies have correlated the concentrations of trace elements with biological (health and reproductive) effects in this group of birds. Most studies have been carried out on specific areas of the Northern Hemisphere, which are supported by monitoring programs that are usually operational.

In gulls, most studies have been performed on feathers and eggs. However, no single study correlated those levels with their levels of internal organs and their possible physiological effects, which would be very useful as a non-invasive proxy for biological effects. On the other hand, similar studies in the Southern Hemisphere are needed.

CONCLUSIONS

It is necessary to overcome the geographic gaps in the availability of information and incorporate monitoring elements that have taken great relevance in recent years, such as the critical elements of technology. The present review compiles the existing worldwide studies of trace elements in species of the *Larus* genus. The compilation and synthesis reported here will be useful for further research on biological monitoring using shorebirds and plans for conserving coastal ecosystems.

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