



Aluminium in aquatic environments: abundance and ecotoxicological impacts

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Abstract Aluminium (Al) is a common chemical element released into the aquatic environment from the Earth's crust and many anthropogenic activities. It may be present in various dissolved and precipitated forms [Al^{3+} , AlOH^{2+} , $\text{Al}(\text{OH})_2^+$, $\text{Al}(\text{OH})_3^0$, $\text{Al}(\text{OH})_4^-$, etc.], which are potentially toxic for organisms. This review summarizes information about the concentrations of Al detected in aquatic ecosystems and its effects on both freshwater and marine organisms (such as growth disturbance, reproduction, and respiration alterations). As the chemistry of Al is different in freshwater and marine systems, we discuss the behaviour of aluminium and its effects on marine or freshwater fauna. Therefore, the solubility of Al, as

other metals, is highly pH dependent, which increases when pH decreases. We are assuming that ocean acidification, linked to climate change, would affect the Al bioavailability in the aquatic environment, which may increase its ecotoxicological effects on semi-closed (Bays, Mediterranean Sea, etc.) or closed (lakes, etc.) aquatic ecosystems.

Keyword Aluminium · Aquatic environment · Effects · Toxicity · Bioavailability

Aluminium (Al) is the third most abundant element in the Earth's crust and is included in class A metal (Nieboer and Richardson 1980; Maya et al. 2016). The erosion of Earth's crust is the main natural Al source, whereas a multitude of anthropogenic activities can explain Al release in the environment (i.e. mineral extraction, industrial activities, etc.). The aqueous chemistry of Al is complex because it can exist in complex forms as many inorganic monomeric species [Al^{3+} , AlOH^{2+} , $\text{Al}(\text{OH})_2^+$, $\text{Al}(\text{OH})_3^0$, and $\text{Al}(\text{OH})_4^-$], amorphous $\text{Al}(\text{OH})_3$, and polynuclear species too (Driscoll and Schecher 1990; Gensemer and Playle 1999; Crane et al. 2007; Millero et al. 2009). Aqueous Al also forms inorganic and organic complexes (Gensemer and Playle 1999). Aluminium accumulation in oceans and seas is underestimated, and the publications mentioning the Al effects on aquatic fauna appear restricted. This metal is abundant

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in aquatic environments; thus, the investigation of its impact is of great importance. Therefore, we decided to write a mini review on the Al abundance and effects on aquatic animals, who involve in many ecological niches (filter, grazer, predator, scavenger, etc.).

The objectives of our review are: (1) to discuss the speciation and solubility of Al in aquatic environments, (2) to do a state of knowledge on Al abundance in aquatic environments, (3) to review Al concentrations found in animal tissues, (4) to summarize the various negative effects on aquatic organisms, and (5) to discuss the potential effect of ocean acidification on the local bioavailability of Al in semi-closed and closed aquatic zones (i.e. bays, lakes, etc.).

Speciation and solubility of aluminium in aquatic environments

Speciation of aluminium

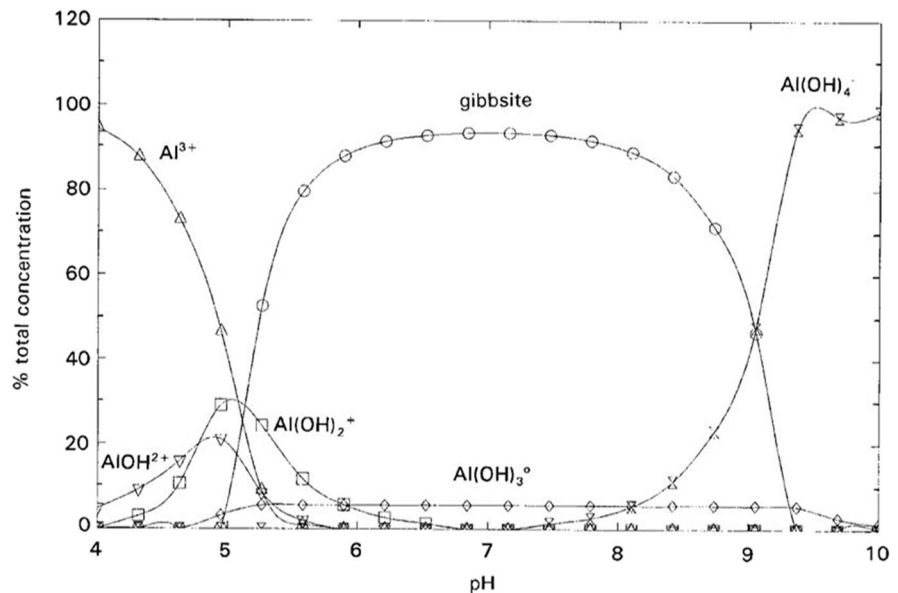
Aluminium can be found in various dissolved or precipitated forms (Gensemer et al. 2018; Angel et al. 2016). Dissolved forms are mainly represented by small species such as $\text{Al}(\text{OH})_4^-$ or $\text{Al}(\text{OH})_3^0$ (Angel et al. 2016; Millero et al. 2009), while precipitated forms are constituted by the transformation of monomeric species of Al into insoluble polymers (Angel et al. 2016; Gensemer et al. 2018). It has been shown that these dissolved and precipitated forms coexist when the solubility threshold is exceeded [e.g. solubility of 0.5 mg/L at 22 °C in neutral pH (8.15) in seawater]. Indeed, when this solubility threshold is exceeded, there is a change in Al speciation where this metal precipitates, mainly in the form of hydroxide $\text{Al}(\text{OH})_3$ or gibbsite (Golding et al. 2015), resulting in a mixture of dissolved and particulate Al (Angel et al. 2016). However, under this solubility limit, Al is mostly present in its dissolved forms (Millero et al. 2009). These dissolved and precipitated forms of Al can both participate in the toxicity of this metal (Golding et al. 2015; Gensemer et al. 2018; Gillmore et al. 2016; Trenfield et al. 2017). Furthermore, after the formation of Al precipitates, these forms change over time (Angel et al. 2016). For example, in seawater, the composition of Al precipitates would change with the incorporation of magnesium forming hydrotalcite (Angel et al. 2016), while they mainly form gibbsite in freshwater environments (Gensemer

and Playle 1999; Santore et al. 2017). However, dissolved and precipitated forms are not the only forms of Al in aquatic environments. Aqueous Al also forms inorganic and organic complexes (Gensemer and Playle 1999) (Figs. 1 and 2). Indeed, Al can form complexes with various inorganic ligands such as fluoride ions (F^-) (Fig. 2B), which can substitute the hydroxide ion (OH^-) due to the similarities of these two ions in terms of charge, ionic radius, and size (Driscoll and Schecher 1990; Nordstrom and May 1996). These complexes are primarily formed under acidic conditions rather than under circumneutral or alkaline conditions. At high pH values, it becomes difficult for ion F^- to compete with hydroxides for Al due to the abundance of OH^- (Driscoll and Schecher 1990). In general, the fluoro and hydroxo complexes remain the primary and strongest inorganic complexes formed with Al (Nordstrom and May 1996; Pyrzyńska et al. 1999). Aluminium can also form AlSO_4^+ and $\text{Al}(\text{SO}_4)_2^-$ by complexing with sulphate, particularly at low pH value, for the same reason as for fluoro complexes (Driscoll and Schecher 1990; Nordstrom and May 1996). The formation of fluoro and sulfato Al complexes depends on pH but also on ionic strength, the concentration of inorganic ligands, and temperature (Gensemer and Playle 1999). Al can also establish complexes with other inorganic ligands such as phosphate and silicate (Gensemer and Playle 1999; Pyrzyńska et al. 1999; Santore et al. 2017). Some organic complexes may also exist with humic and fulvic acids (Fig. 2C) (Pyrzyńska et al. 1999; Driscoll and Schecher 1990).

Environmental factors modulating the Al solubility

Water quality factors are important to consider because they can affect the chemical speciation of Al, which plays a key role in the bioavailability and toxicity of this metal (Gabelle et al. 2012; Santore et al. 2017; Gensemer et al. 2018). Al solubility in aquatic systems is dependent on several chemical factors and, in particular, on pH, dissolved organic carbon (DOC) concentration, salinity (i.e. cations), and temperature (Santore et al. 2019). Al is mostly insoluble within the range of pH 6 to 8 (Fig. 1). Thus, within the pH range of most natural surface waters, Al primarily exists in its insoluble form $\text{Al}(\text{OH})_3$ or gibbsite (Wren and Stephenson, 1991; Gensemer et al.

Fig. 1 Aluminum speciation from a MINEQL⁺ simulation (Schecher and McAvory, 1992) using five aquatic species of Al plus gibbsite, and varying water pH from pH 4 to 10. The simulation was run using 4 μM total Al (=100%) and 15°C. In the simulation, we used 1 mM concentrations for Ca, Cl, Na, and NO₃, with the system open to the atmosphere (Gensemer and Playle 1999)



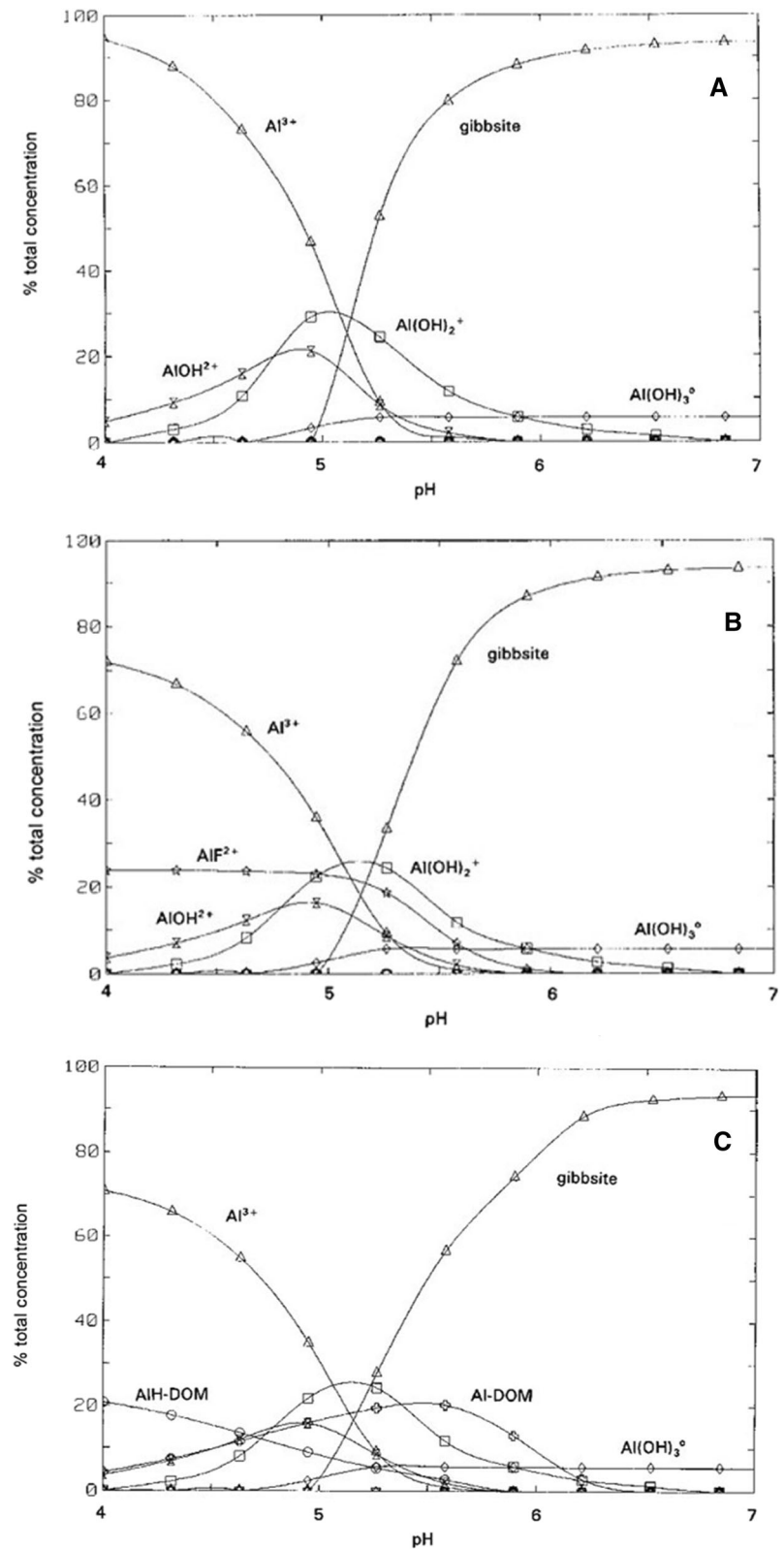
2018; Zhou et al. 2018) (Fig. 1). The solubility of Al is increased in acidic conditions (pH < 6.0), where its prevalent forms are the free ion Al³⁺ (which is better indicator to metal toxicity, Campbell et al. 1995) and inorganic monomeric species. In alkaline conditions (pH > 8.0), the predominant species are inorganic monomeric species Al(OH)₄⁻ (Fig. 1) (Driscoll and Schecher 1990; Wren and Stephenson, 1991; Crane et al. 2007; Millero et al. 2009). Thus, aluminium becomes more soluble and its toxicity increases at low and high pH (Driscoll and Schecher 1990; Gensemer and Playle 1999; Santore et al. 2017). Moreover, hardness of water also plays a role in the speciation of Al (Gensemer and Playle 1999). In fact, hardness may have a protective effect on Al toxicity (Gensemer et al. 2018). Moreover, water temperature is also an important factor to consider because it affects the solubility, hydrolysis, and speciation of Al in aquatic environments (Lydersen 1990; Gensemer and Playle 1999). For instance, Lydersen et al. (1990) showed that polymerization processes of Al would be more advanced at high temperatures than at low temperatures. Lydersen (1990) showed that at 25 °C (pH 5), 36% of Al is transformed into Al³⁺, whereas at 0 °C (pH 5) this form represents 86% of Al. Furthermore, DOC concentration is also an important factor concerning the solubility of Al (Santore et al. 2017). Indeed, when Al is bound to dissolved organic matter (DOM), its solubility increases, and its toxicity

decreases (Gensemer and Playle 1999; Gabelle et al. 2012; Santore et al. 2017).

Aluminium abundance in aquatic ecosystems

The weathering of rocks and minerals allows a sediment deposition of Al along rivers, estuaries, and coastal waters, whereas offshore and open ocean locations depend on river runoff (Zhou et al. 2018). It can also be abundantly released by anthropogenic activities (like air emissions, wastewater effluents, and solid waste) mainly associated with industrial processes (Jones et al. 2005; Hadi et al. 2009). For instance, water treatment systems use Al salts (aluminium sulphate Al₂(SO₄)₃ and aluminium chloride AlCl₃), which are subsequently found in natural waters (Takaoka et al., 2015). Another potential anthropogenic source is the use of Al associated with metals (indium, manganese, zinc) as sacrificial anodes materials to protect immersed steel structures from corrosion in ports (Leleyter et al. 2018). Aluminium alloy is found to delay corrosion, involving a progressive release and accumulation of Al in water and especially surrounding sediments (Gabelle et al. 2012; Leleyter et al. 2018). Furthermore, in freshwater, Al concentrations can be increased in response to lake acidification and acid precipitations, which can induce the transport of Al through soil to streams, lakes, and

Fig. 2 **A** Aluminum speciation from MINEQL⁺ using five aquatic species of Al plus gibbsite, and varying water pH from pH 4 to 7. Total Al = 4 μM . See Figure caption 1 for more details of the simulation. **B** The same simulation as in 3A, with the addition of 1 μM F^- . **C** The same simulation as in 3A, with the addition of two Al-DOM species into the MINEQL⁺ program, AlH-DOM (log K = 13.1) and Al-DOM (log K = 8.4). The DOM species total 1 μM in the simulation (about 0.3 mg C · L⁻¹ DOC). See text for more details. The AlOH^{2+} and $\text{Al}(\text{OH})_2^+$ species have the same symbols as in 3A and 3B (Gensemer and Playle 1999).



ground waters (Cronan and Schofield 1979). Indeed, the decrease in pH in water and soil increases the mobility of Al in the environment (Pyrzynska et al. 1999). Thus, with acid leaching, the mobilization of Al in soil can occur, resulting in the transport of Al from edaphic to aquatic environments (Driscoll and Schecher 1990). Table 1 summarizes dissolved Al concentration values in the marine and freshwater environments in various regions of the world, covering a relatively large range of Al concentrations.

Aluminium in freshwater

Some measurements of dissolved Al concentrations in freshwater environments showed values ranged from 0.324 to 928 µg/L in rivers and from 120 to 3700 µg/L in lakes (Table 1). These freshwater locations are affected by anthropogenic activities, like in the Weihe River (China) running through Xi'an, which is the biggest city in northwestern China (Wang et al. 2013). Indeed, industrial activities that take place in Xi'an affect the quality of the river water, which presents high levels of dissolved Al. Therefore, high concentrations of dissolved Al are also released into the effluents of the Weihe River mostly by papermaking factories (which use Al salts to treat wastewaters) (Wang et al. 2013). Furthermore, it has been shown that dissolved Al concentrations may also depend on the amount of DOM which increases the Al solubility (Gensemer and Playle 1999; Gabelle et al. 2012; Santore et al. 2017), as suggested by van Bennekom and Jager (1978), for the Zaire River (the second longest river in Africa). Upadhyay et al. (2002) also indicated that DOM, but also pH, could play an important role in the distributions of dissolved Al in the Yare River and the Great Ouse River (England). A low pH can also partly explain the high dissolved Al concentrations found in the two Japanese lakes: the Lake Usoriko (pH = 3.0–3.6) and the Lake Inawashiroko (pH = 5.1–5.2). In fact, these lakes have the particularity to be acidic because they are partly supplied by acidic waters of volcanic origins and an acidic stream (Takatsu et al. 2000). However, as said earlier, Al solubility is increased in acidic conditions, which could increase its bioavailability and thus increase its toxicity. Takatsu et al. (2000) showed that in the Lake Usoriko the dominant form of Al is Al³⁺, whereas in the Lake Inawashiroko it is the form Al combined with a ligand that is dominant. Moreover,

the bioaccumulation factor of Al of the fish *Tribolodon hakonensis*, adapted to this acidic environment, was lower in the Lake Usoriko than in the Lake Inawashiroko (Takatsu et al. 2000). Thus, even if the pH is lower, the uptake seems not to be higher. Furthermore, the form Al³⁺ may not be the most bioavailable form of Al.

Based upon a species sensitivity distribution of freshwater organisms exposed to Al in toxicity tests, Cardwell et al. (2018) defined a generic (not corrected for water chemistry or bioavailability), Al concentration of 74.4 µg total Al/L for protection of freshwater organisms was used in toxicity testing. For example, Baudouin et al (2008) mentioned Al concentration in streams in the Vosges Mountains and found total Al concentrations ranging from 49 to 652 µg/L, so exceeded the value defined by Cardwell et al. (2018), meaning that in these streams Al total concentration can be toxic to freshwater organisms. Unlike the dissolved Al concentrations measured in aquatic environments (Table 1), the Al concentration defined by Cardwell et al. (2018) is based on total Al and not dissolved Al. However, dissolved and precipitated forms of Al can both participate in its toxicity (Golding et al. 2015; Gensemer et al. 2018; Gillmore et al. 2016; Trenfield et al. 2017). Thus, we cannot really estimate the potential Al toxicity based on dissolved Al concentrations in these environments. Furthermore, this theoretical protective concentration is exceeded by some of the values of Al concentrations reported in the literature, such as for the Weihe River (China) (Wang et al. 2013), the Selenga River (Mongolia) (Myangan et al. 2017), the lake Usoriko (Japan), or the lake Inawashiroko (Japan) (Takatsu et al. 2000). Even if these surface water concentrations are based upon the dissolved forms of Al, there is already a potential ecotoxicological risk for freshwater organisms at these locations. Thus, several biological processes of these freshwater organisms, such as their reproduction or their growth, may already have been affected by current Al concentrations. One exception is the annelid *Aeolosoma sp.*, which has an EC₁₀ of 987.9 µg total Al/L (Cardwell et al. 2018), meaning that its population size might not be reduced by the higher current Al concentration measured in rivers (928 µg dissolved Al/L; Table 1). Therefore, environmental concentrations of dissolved Al could not affect this freshwater organism according to water parameters. However, precipitated forms of Al can induce

Table 1 Concentrations of dissolved Al ($\mu\text{g/L}$) in freshwater, coastal, and open ocean waters

Locations	Method used	Dissolved Al ($\mu\text{g/L}$)	References
<i>Freshwater locations</i>			
Weihe River, Xi'an section, China	AAS	65–928	Wang et al. 2013
Selenga River and its tributaries, Mongolia	ICP-AES	10–100	Myangan et al. 2017
Malela, Zaire River, Congo	AAS	28–44	van Bennekom and Jager 1978
Asaba, Niger River, West Africa	AAS	3–6	van Bennekom and Jager 1978
Columbia River, USA	ICP-MS	2.16	Brown and Bruland 2009
Tenryu River, Japan	ICP-AES	20	Takatsu et al. 2000
Yare River, England	FS1	0.324–2.75	Upadhyay et al. 2002
Great Ouse River, England	FS1	0.864–7.4	Upadhyay et al. 2002
Lake Usoriko, Japan	ICP-AES	510–3700	Takatsu et al. 2000
Lake Inawashiroko, Japan	ICP-AES	120–300	Takatsu et al. 2000
<i>Coastal waters</i>			
Al-Khobar coastline, Arabian Gulf, Saudi Arabia	ICP-MS	0.60–10.60	Alharbi et al. 2017
Al-Khafji coastline, Arabian Gulf, Saudi Arabia	ICP-MS	0.68–1.56	Alharbi and El-Sorogy 2019
Arabian sea, Indian coast	FS2	1.49–2	Upadhyay and Sen Gupta 1994
Marsa Matrouh Beaches, Egypt	AAS	19.25–106.49	Ghani 2015
Florida Bay, USA	ICP-MS	0.81–48.5	Caccia and Millero 2003
Northern Taiwan coast	FS2	1.81–3.55	Fang et al. 2006
San Jorge Bay, Antofagasta, northern Chile	HHPM-AAS	67.42–131.96	Valdes et al. 2011
Outer Harbour, Port Curtis, Queensland, Australia	ICP-MS	4–5	Angel et al. 2012
Outside Port Curtis, Queensland, Australia	ICP-MS	2–4	Angel et al. 2012
Cronulla, NSW, Australia	ICP-AES	1.3	Angel et al. 2016
Black Sea in Rize, Turkey	ICP-OES	33–80	Baltas et al. 2017
Gulf of Guinea, Africa	AAS	0.8 ± 0.3	van Bennekom and Jager 1978
<i>Open Ocean locations</i>			
Northwestern Indian Ocean, Arabian Sea	FS2	0.99–1.4	Upadhyay and Sen Gupta 1994
Southern Ocean (Weddell Sea)	AAS	$0.085 (\pm 0.035)$	Sañudo-Wilhelmy et al. 2002
Central Arctic ocean	FS2	0.111–0.567	Moore 1981
North East Atlantic	FS2	0.432–0.864	Hydes 1983
North Atlantic Ocean	FS2	0.165–0.689	Kramer et al. 2004
Mediterranean, eastern basin	FS2	1.49–2.03	Hydes et al. 1988

AAS, Atomic absorption spectrophotometry, *FS1*, fluorescence spectrometry 1: Upadhyay and Sen Gupta, 1994; *FS2*, fluorescence spectrometry 2: Hydes and Liss, 1976; *HHPN-AAS*, hydraulic high-pressure nebulization flame furnace atomic absorption spectrometry; *ICP-AES*, inductively coupled plasma atomic emission spectroscopy; *ICP-OES*, inductively coupled plasma optical emission spectrometry; *ICP-MS*, inductively coupled plasma mass spectroscopy

toxicity, especially for fish, as it has been suggested by Poléo (1995) and Alexopoulos et al. (2003). Therefore, the already potentially dangerous environmental dissolved Al concentrations can be more harmful to freshwater organisms.

The United States Environmental Protection Agency published in 1988 freshwater criteria for Al to protect aquatic life from harmful effects of its

toxicity (750 $\mu\text{g/L}$ of recoverable Al after 1 h of exposure). These criteria were re-evaluated in 2018, and it is now 800 $\mu\text{g/L}$. After 4 days of exposure, it is 200 $\mu\text{g/L}$. It depends on pH's sites, total hardness, and DOC (US EPA 2018). In Europe, there is now a WHO guideline value (from 2011) for Al in drinking water from water treatment plants of 0.1 mg/L in large treatment plants and 0.2 mg/l in small treatment

plants, based on pragmatic optimization of the treatment process. This guide value is set primarily for reasons of taste and appearance (WHO 2011). If we compare these regulations to the freshwater Al concentrations (Table 1), we can see that two of the locations have a dissolved Al concentration higher: the Weihe River (China), which is highly contaminated by anthropogenic activities (Wang et al. 2013), and the Lake Usoriko (Japan), which has a low pH that could explain a high rate of dissolved Al (Takatsu et al. 2000).

Aluminium in seawater

Dissolved Al concentrations measured in coastal waters ranged from 0.6 to 131.96 $\mu\text{g/L}$, while the concentrations in the open ocean are between 0.09 and 2.03 $\mu\text{g/L}$.

The fact that coastal waters have higher concentrations of Al over open ocean locations could be accounted for by greater exposure to multiple anthropogenic sources, impacting mostly coastal locations (Gabelle et al. 2012; Leleyter et al. 2018). For example, the highest dissolved Al concentration was observed in the northern coast of Chile, which is highly densely populated including the port city of Antofagasta (the most important human settlement in northern Chile) (Valdes et al. 2011). Indeed, the San Jorge Bay is subject to economic and industrial activities but also to the wastes of old mining activities (Valdes et al. 2011). Other places showed high concentrations of dissolved Al, like the Egyptian coast (Ghani 2015) and the port Curtis (Australia) (Angel et al. 2012), for which several potential sources of Al exist, such as alumina refineries and an Al smelter (Angel et al. 2012, 2016). On the northern coast of Taiwan, sources of pollution may be due to domestic dumps and industrial effluents (Fang et al. 2006). The high measurements of dissolved Al concentrations in the Arabian Gulf (Sites: Al-Khobar and the Al-Khafji coastlines) could be due to fluvial sources and possibly the mobilization of Al from sediments or pore waters (Upadhyay and Sen Gupta 1994; Alharbi et al. 2017; Alharbi and El-Sorogy 2019). These two areas are affected by anthropogenic activities such as landfilling, desalination factories, atmospheric inputs, or oil spills (Alharbi et al. 2017; Alharbi and El-Sorogy 2019). Alharbi et al. (2017) also identified fishing boats and rubbish on the

coastline as sources of metal pollution in the Al-Khobar coastline. Florida Bay (USA), another coastline location, contained the lowest levels of dissolved Al measured in the South-central zone and the highest measured in the North-central and Western zones (Caccia and Millero 2003). Caccia and Millero (2003) explained these differences by the influence of precipitations and river runoff. Indeed, Al could have been transported by the waters of the Gulf of Mexico from the rivers of Ten Thousand Islands subject to agricultural activities, and especially from the Shark River, which is considered by these authors as a major source of metals. Thus, the low concentrations of Al measured in the South-central zone of the Florida Bay could be explained by the dilution of metals due to the waters of the Gulf Stream and the Atlantic ocean through the channels along the Florida Keys (Caccia and Millero 2003).

The open ocean locations showed a range of dissolved Al concentrations lower than coastal sites (Table 1). The lowest concentration has been measured in the Weddell Sea (Part of the South Atlantic Ocean near the Antarctic), which should be less affected by anthropogenic pressures than the other locations. The major sources of dissolved Al in the open ocean locations are often described as atmospheric inputs (Hydes 1983; Hydes et al. 1988; Upadhyay and Sen Gupta 1994; Kramer et al. 2004). For example, Al concentration measured in the North Atlantic Ocean could depend on Saharan dust (Hydes 1983; Kramer et al. 2004). Moreover, the Mediterranean Sea contained more dissolved Al than the other open locations (Table 1) and is also subject to dust inputs (Hydes et al. 1988).

As for freshwater organisms, a water quality guideline value has been established for marine organisms using the data of studies, which realized chronic toxicity tests and have been described as acceptable (Van Dam et al. 2018). Indeed, Van Dam et al. (2018) recommended a water quality guideline value for dissolved Al in seawater of 56 $\mu\text{g/L}$ to protect 95% of marine species. However, this value has been established using tropical and temperate marine species (Van Dam et al. 2018) and thus creates uncertainty in its validity for all marine organisms. No water quality guideline seems to have been established for Al concerning only temperate or arctic species. Even though the concentrations measured by Ghani (2015) and Baltas et al. (2017) in Mediterranean coasts

(Egypt), or Valdes et al. (2011) along the Chilean Bay only report dissolved Al, these concentrations are already above the water quality guideline value given by van Dam et al. (2018) and may thus indicate an ecotoxicological risk for the marine fauna in these locations. In fact, it has been established that, at neutral pH, the precipitated and dissolved forms of Al can both participate in its toxicity (Golding et al. 2015; Gensemer et al. 2018; Gillmore et al. 2016; Trenfield et al. 2017). Another water guideline value has been established by Golding et al. (2015), but this value was determined solely using chronic toxicity tests with Australian species. However, this study revealed that the total Al concentration should not exceed 24 $\mu\text{g/L}$ to protect 95% of the marine tropical species (Golding et al. 2015). An important difference between the two water quality guideline values cited is the number or identity of species used. The study of Golding et al. (2015) included 11 Australian species, while van Dam et al. (2018) based their value on 17 species found in the literature for tropical and temperate zones. Therefore, the value established by van Dam et al. (2018) could be more accurate considering the potential effects that temperature could have on Al speciation (Lydersen 1990; Gensemer and Playle 1999) and thus its toxicity.

Aluminium abundance in aquatic animals

Al concentrations in animal tissues have been measured in freshwater and marine organisms, and some values are presented in Tables 2, 3, and 4. This Al content is measured as dry weight of organisms (Beiras et al. 2003; Slaninova et al. 2014; Duysak and Azdural 2017; Primost et al. 2017), or as wet weight (Ranau et al. 2001; Ghani 2015; Baltas et al. 2017).

In marine organisms, Al concentrations in tissues and organs are heterogeneous, particularly for mollusks, for which the tissues can be the digestive gland and the gonad complex (Primost et al. 2017), the foot (Primost et al. 2017), the soft tissues (Beiras et al. 2003), or the gill tissue (Duysak and Azdural 2017). However, Al concentrations can be very different according to the tissue considered. For example, Al concentrations in the foot of *Buccinanops globulosus* ranged from 21.39 to 103.32 $\mu\text{g/g}$ dry weight, while the values for the digestive gland and the gonad complex ranged from 13.31 to 17.21 $\mu\text{g/g}$ dry weight

(Primost et al. 2017). These heterogeneities in the data make direct comparisons difficult. Tables 2 and 4 show that Al concentrations measured in fish species muscles ranged from 0.078 to 100 $\mu\text{g/g}$ wet weight in coastal waters (Ranau et al. 2001; Ghani 2015; Baltas et al. 2017), while in open ocean locations, these concentrations are between 0.03 and 0.295 $\mu\text{g/g}$ wet weight (Ranau et al. 2001). The coastal fish may be more exposed to Al than open ocean species, possibly because the littoral zones are close to Al emissions caused by many anthropogenic activities. Al concentrations measured in the same location in fish, mollusks, and arthropods show important differences, with higher Al concentrations in the molluscan and arthropod tissues (Table 5). For example, in the North Sea, Al concentrations ranged from 0.205 to 4.95 $\mu\text{g/g}$ wet weight for mollusks and from 2.41 to 3.392 $\mu\text{g/g}$ wet weight for arthropods, while they ranged from 0.046 to 0.159 $\mu\text{g/g}$ wet weight for fish (Ranau et al. 2001). This could be due to a higher accumulation of Al in marine shellfish species because they are filter feeders, which are more exposed to metals in their environment (Ranau et al. 2001). Multiple Al concentrations in tissues of marine organisms are listed in Tables 3, 4, and 5.

Concerning freshwater organisms, Wren and Stephenson (1991) showed in review various Al concentrations estimated in invertebrates (Cladocera, Decapoda, Mollusca, and Insecta), indicating high Al contents in zooplankton (263–358 $\mu\text{g/g}$ ww) and in insects (757–2130 $\mu\text{g/g}$ ww). Kuklina et al. (2014) compared the accumulation of Al in three tissues (gills, hepatopancreas, and abdominal muscle) in the crayfish *Astacus astacus* between four water supply reservoirs in the Czech Republic, one being known to be contaminated (Darkovske) and the three others not contaminated (Boskovice, Landstejn, and Nova Rise). It appears that for this crayfish, gills are the organs accumulating most of the Al compared to the hepatopancreas and the abdominal muscle. Therefore, the choice of tissue in biomonitoring studies concerning Al and other metals is important to not over- or underestimate the Al concentration in the environment (Kuklina et al. 2014). Walton et al. (2009) investigated in the laboratory the possible link between Al accumulation and its toxicity, and more precisely its toxicity on the activity of the freshwater gastropod *Lymnaea stagnalis*. They found out that Al tissue content did not allow the prediction of Al toxicity in

Table 2 Al concentrations ($\mu\text{g/g}$ wet weight) in tissues of marine organisms in coastal waters

Location	Phylum	Species	Tissue	Al concentration ($\mu\text{g/g}$ wet wt)	Range ($\mu\text{g/g}$ wet wt)	References
Marsa Matrouh Beaches, North-western Mediterranean coast, Egypt	Chordata	<i>Saurida undosquamis</i>	Muscle	12.011		Ghani (2015)
		<i>Boops boops</i>		14.239		
		<i>Adioryx diadema</i>		8.291		
		<i>Pagellus erythrinus</i>		100.000		
		<i>Mullus surmuletus</i>		10.895		
		<i>Diplodus sargus</i>		19.890		
		<i>Sparisoma cretense</i>		16.876		
		<i>Dicentrarchus labrax</i>		1.784		
Rize shore, Black Sea, Turkey	Chordata	<i>Engraulis encrasicolus</i>	Soft tissue		3.937–6.307	Baltas et al. (2017)
Stavanger/Norway, near Skudeneshavn	Chordata	<i>Gadus morhua</i>	Muscle	0.131	0.129–0.133	Ranau et al. (2001)
		<i>Pollachius pollachius</i>		0.091	0.082–0.1	
		<i>Pollachius virens</i>		0.082	0.068–0.095	
		<i>Melanogrammus aeglefinus</i>		0.151	0.070–0.226	
		<i>Merlangius merlangus</i>		0.134	0.074–0.221	
		<i>Molva molva</i>		0.099	0.081–0.118	
		<i>Brosme brosme</i>		0.108	0.081–0.128	
		<i>Scomber scombrus</i> L.		0.101	0.035–0.2	
		<i>Anarhichas lupus</i>		0.225		
		<i>Limanda limanda</i>		0.128	0.11–0.145	
		Stavanger/Norway, Akrehamn		Chordata	<i>Gadus morhua</i>	
<i>Pollachius pollachius</i>	0.281		0.195–0.446			
<i>Melanogrammus aeglefinus</i>	0.936		0.664–1.232			
<i>Molva molva</i>	0.285		0.271–0.304			
<i>Scomber scombrus</i> L.	0.078		0.057–0.098			
<i>Belone belone</i>	0.161		0.159–0.162			

this gastropod. Slaninova et al (2014) mentioned that Al concentrations ranged from 13.3 ± 7.85 to 23.5 ± 8.55 $\mu\text{g/g}$ dry matter in the fish *Cyprinus carpio* collected in the Czech Republic.

Negative effects of aluminium in aquatic organisms

Several studies assessed the negative effects of Al on marine and freshwater fauna. Table 5 presents the effects of acute toxicity but also the effects of chronic

Table 3 Al concentrations ($\mu\text{g/g}$ dry weight) in tissues of organisms in coastal waters

Location	Phylum	Species	Tissues	Al concentration ($\mu\text{g/g}$ dry wt)	Range ($\mu\text{g/g}$ dry wt)	References
Galician Rias of Vigo, Pontevedra, and Arousa	Mollusca	<i>Mytilus galloprovincialis</i>	Soft tissues		168–1237	Beiras et al. (2003)
Nuevo Gulf, Argentina	Mollusca	<i>Adelomelon ancilla</i>	Foot	23.2		Primost et al. (2017)
			Digestive gland-gonad complex	13.04		
		<i>Buccinanops globulosus</i>	Foot		21.39–103.32	
			Digestive gland-gonad complex		13.31–17.21	
		<i>Trophon geversianus</i>	Foot		54.01–71.89	
			Digestive gland-gonad complex	15.55		
Iskenderun Bay, Turkey	Mollusca	<i>Patella caerulea</i>	Gill tissue		35.4–157.8	Duysak & Azdural (2017)
			Liver tissues		15.3–97.7	
			Muscular tissue		16.5–53.0	

toxicity to Al, which is more representative of the effects of long-term exposure that organisms and populations can encounter in their environment.

Aluminium effects in freshwater organisms

The toxicity of Al in freshwater species has been studied extensively in fish. It has been shown that, at circumneutral pH conditions (pH = 6.0–8.0), negative respiratory effects predominate which could be due to the accumulation of precipitated forms of Al on the gill surface (Gensemer and Playle 1999; Gensemer et al. 2018). Aluminium polymerization on the fish gills probably causes a physical surface effect of clogging, leading to hypoxia in addition to osmoregulation dysfunction (Poléo 1995; Alexopoulos et al. 2003). The clogging of the interlamellar spaces and the increase in mucus secretion will result in the reduction in water flow over the respiratory surfaces and an increased thickness of the diffusion barrier for gases and ions (Poléo 1995). In addition, to these mechanisms, Al causes histopathological changes in the liver (loss of cytoplasm, loss of centrilobular, loss of central canal, loss of hepatic cell, cytoplasmic

vacuolation, necrosis of hepatic cells) and kidney (expansion of Bowman's loss of renal tubules, narrowing of tubular lumen, damage of hematopoietic cells, clustering of hemosiderin pigments, clustering of kidney cells, and glomerulus vacuolation) of the freshwater fish *Clarias batrachus* in neutral condition (pH = 7.0) (Raj et al. 2018). Correia et al. (2010) showed that Al could also be considered as an endocrine disruptor for females of the fish species *Oreochromis niloticus* at neutral pH.

Some studies were also carried out on freshwater invertebrates indicating reproduction impairment and mortality (Wren and Stephenson 1991). The publications on mollusks showed an interesting mechanism of Al regulation by the synthesis of metallic granules. For instance, it has been shown that freshwater snail *Lymnaea stagnalis* is capable of accumulating Al at neutral pH in its soft tissues and mainly in its digestive glands (which would act as a sink for Al), and in its kidneys (which would be a regulatory site) (Elangovan et al. 1997). Indeed, the role of the digestive gland is particularly due to the presence of excretory “granules”, which are able to accumulate and probably detoxify Al and have been observed in *L. stagnalis*

Table 4 Al concentrations ($\mu\text{g/g}$ wet weight) in tissues of marine organisms in open sea locations

Location	Phylum	Species	Tissue	Al concentration ($\mu\text{g/g}$ wet wt)	Range ($\mu\text{g/kg}$ wet wt)	References		
North Sea	Chordata	<i>Gadus morhua</i>	Muscle	0.076	0.033–0.192	Ranau et al. (2001)		
		<i>Pollachius pollachius</i>		0.046	0.021–0.089			
		<i>Pollachius virens</i>		0.099	0.032–0.155			
		<i>Melanogrammus aeglefinus</i>		0.162	0.034–0.349			
		<i>Merlangius merlangus</i>		0.069	0.067–0.07			
		<i>Molva molva</i>		0.067	0.037–0.18			
		<i>Merluccius merluccius</i>		0.056	0.025–0.088			
		<i>Lophius piscatorius</i>		0.078	0.032–0.131			
		<i>Zeus faber L.</i>		0.101				
		<i>Scomber scombrus L.</i>		0.074	0.04–0.102			
		<i>Trachurus trachurus L.</i>		0.102	0.057–0.149			
		<i>Engraulis encrasicolus L.</i>		0.159				
		<i>Hyperoplus lanceolatus</i>		0.086				
		<i>Conger conger</i>		0.080	0.064–0.105			
		<i>Anarhichas lupus</i>		0.052	0.033–0.094			
		<i>Pleuronectes platessa</i>		0.102	0.048–0.176			
		<i>Limanda limanda</i>		0.11	0.061–0.176			
		<i>Platichthys flesus</i>		0.101	0.088–0.113			
		<i>Microstomus kitt</i>		0.064	0.04–0.111			
		<i>Solea vulgaris</i>		0.065	0.056–0.073			
		<i>Glyptocephalus cynoglossus</i>		0.108				
		<i>Psetta maxima</i>		0.088	0.042–0.177			
		<i>Hippoglossus hippoglossus</i>		0.095				
		Arthropoda		<i>Nephrops norvegicus</i>	Abdominal muscle		2.41	1.538–3.73
				<i>Cancer pagurus L.</i>			3.392	2.162–5.434
		Mollusca		<i>Loligo spp.</i>	Tubes and tentacles		0.205	0.111–0.352
		<i>Buccinum undatum</i>	Muscle	4.067	2.023–6.202			
		<i>Mytilus edulis</i>		4.95	3.793–6.107			

Table 4 continued

Location	Phylum	Species	Tissue	AI concentration ($\mu\text{g/g}$ wet wt)	Range ($\mu\text{g/kg}$ wet wt)	References		
Northeast Atlantic	Chordata	<i>Pollachius pollachius</i>	Muscle	0.042				
		<i>Pollachius virens</i>		0.058	0.026–0.093			
		<i>Melanogrammus aeglefinus</i>		0.132				
		<i>Merlangius merlangus</i>		0.088	0.033–0.164			
		<i>Micromesistius poutassou</i>		0.058	0.051–0.071			
		<i>Molva dipterygia</i>		0.051	0.041–0.076			
		<i>Brosme brosme</i>		0.1	0.084–0.14			
		<i>Phycis blennoides</i>		0.055	0.051–0.058			
		<i>Merluccius merluccius</i>		0.064	0.042–0.091			
		<i>Coryphaenoides rupestris</i>		0.127	0.09–0.178			
		<i>Lophius piscatorius</i>		0.052	0.047–0.062			
		<i>Argentina silus</i>		0.091	0.075–0.114			
		<i>Chimaera monstrosa</i>		0.083				
		<i>Squalus acanthias</i> L.		0.087	0.055–0.105			
		<i>Clupea harengus</i>		0.132	0.098–0.173			
		<i>Sebastes</i> spp.		0.096	0.065–0.125			
		<i>Helicolenus dactylopterus</i>		0.062				
		<i>Pleuronectes platessa</i>		0.069	0.045–0.089			
		<i>Limanda limanda</i>		0.073	0.054–0.083			
		<i>Lepidorhombus whiffiagonis</i>		0.087	0.064–0.1			
		<i>Microstomus kitt</i>		0.054	0.035–0.067			
		<i>Glyptocephalus cynoglossus</i>		0.085	0.039–0.131			
		<i>Hippoglossoides platessoides</i>		0.074				
		<i>Reinhardtius hippoglossoides</i>		0.138	0.058–0.272			
		<i>Hippoglossus hippoglossus</i>		0.03	0.021–0.036			
		Arthropoda		<i>Pandalus borealis</i>	Abdominal muscle	1.253	0.941–1.565	
		Mollusca		<i>Loligo</i> spp.	Tubes and tentacles	0.21		
<i>Eledone cirrosa</i>				0.247				

Table 4 continued

Location	Phylum	Species	Tissue	AI concentration ($\mu\text{g/g}$ wet wt)	Range ($\mu\text{g/kg}$ wet wt)	References				
Barents Sea	Chordata	<i>Gadus morhua</i>	Muscle	0.061	0.048–0.075					
		<i>Boreogadus saida</i>		0.191						
		<i>Melanogrammus aeglefinus</i>		0.051	0.038–0.063					
		<i>Micromesistius poutassou</i>		0.102	0.094–0.11					
		<i>Mallotus villosus</i>		0.272	0.233–0.311					
		<i>Sebastes spp.</i>		0.115	0.059–0.147					
		<i>Anarhichas lupus</i>		0.094	0.05–0.135					
		<i>Anarhichas minor</i>		0.055	0.031–0.097					
		<i>Hippoglossoides platessoides</i>		0.076	0.049–0.095					
				Arthropoda	<i>Pandalus borealis</i>		Abdominal muscle	1.013	0.769–1.256	
	Mollusca	<i>Eledone cirrosa</i>	Tubes and tentacles	1.312	1.184–1.44					
Greenland waters	Chordata	<i>Molva dipterygia</i>		0.043	0.037–0.048					
		<i>Brosme brosme</i>		0.056						
		<i>Sebastes spp.</i>		0.069	0.041–0.136					
		<i>Anarhichas lupus</i>		0.053	0.044–0.067					
		<i>Anarhichas minor</i>		0.064	0.057–0.07					
		<i>Reinhardtius hippoglossoides</i>		0.118	0.092–0.15					
		<i>Hippoglossus hippoglossus</i>		0.062	0.037–0.083					
				Arthropoda	<i>Pandalus borealis</i>		Abdominal muscle	1.021	0.96–1.082	
		Baltic Sea		Chordata	<i>Gadus morhua</i>			0.111	0.055–0.195	
					<i>Merlangius merlangus</i>			0.144		
<i>Gadus morhua</i>	0.295		0.236–0.330							
<i>Scomber scombrus L</i>	0.117		0.094–0.155							
<i>Clupea harengus</i>	0.11		0.072–0.147							
<i>Engraulis encrasicolus L.</i>	0.182									
<i>Sprattus sprattus</i>	0.173		0.114–0.212							
<i>Limanda limanda</i>	0.166		0.11–0.250							
<i>Platichthys flesus</i>	0.158		0.088–0.251							
<i>Psetta maxima</i>	0.17		0.142–0.191							
<i>Pleuronectes platessa</i>	0.128		0.115–0.141							

Table 5 Aluminium effects on various aquatic organisms

Type of organism	Phylum	Tested species	Tested endpoint	Test duration	Temperature	pH	Hardness (mg/L CaCO ₃)	Toxicity measure ^b	Al concentration (µg/L)	Range (µgAl/L)	References
Freshwater species											
	Annelida	<i>Aeolosoma</i> sp.	Population size reduction	17 days	24–25°C	6.0	48	EC10	987.9	336.6–2899.4	Cardwell et al. (2018)
	Rotifera	<i>Brachionus calyciflorus</i>	Population growth rate reduction	48h	25°C	6.1–6.5	100	EC10	441.2	284.8–683.5	Cardwell et al. (2018)
	Arthropoda	<i>Ceriodaphnia dubia</i>	Fecundity	7 days	20°C	6.3	25	EC10 ^c	281	191–414	Gensemer et al. (2018)
		<i>Hyalella azteca</i>	Reproduction reduction	42 days	22–25°C	6.0	95	EC10	170.6	126.8–229.6	Cardwell et al. (2018)
		<i>Hyalella azteca</i>	Growth (biomass) reduction	28 days	22–25°C	6.0	95	EC10	142.6	66.8–304.2	Cardwell et al. (2018)
		<i>Hyalella azteca</i>	Growth (biomass) reduction	42 days	22–25°C	6.0	95	EC10	227.7	59.2–875.9	Cardwell et al. (2018)
	Mollusca	<i>Lymnaea stagnalis</i>	Growth (wet weight) reduction	30 days	25°C	6.0–6.1	117	EC10	860.7	733.1–1010.5	Cardwell et al. (2018)
	Chordata	<i>Danio rerio</i>	Growth (biomass) reduction	33 days	24–26°C	5.9–6.3	83	EC10	98.2	33.0–292.2	Cardwell et al. (2018)
		<i>Pimephales promelas</i>	Growth (biomass) reduction	33 days	25 °C	5.9–6.5	96	EC10	417.4	150.6–1157.0	Cardwell et al. (2018)

Table 5 continued

Type of organism	Phylum	Tested species	Tested endpoint	Test duration	Temperature	pH	Hardness (mg/L CaCO ₃)	Toxicity measure ^b	AI concentration (µg/L)	Range (µgAI/L)	References	
Marine species	Ochrophyta	<i>Ecklonia radiata</i>	Germination success	72h	21 °C	8.2	NS	IC10	6800	5400–8200	Golding et al. (2015)	
		<i>Ceratoneis closterium</i>	Growth rate inhibition	72h	21 °C	8.2	NS	IC10	80	55–100	Gillmore et al. (2016)	
		<i>Ceratoneis closterium</i>	Growth rate inhibition	72h	21 °C	8.2	NS	IC10	16	3.2–78	Golding et al. (2015)	
			<i>Nitzschia closterium</i>	Growth rate inhibition	72h	32 °C	8.2	80–110	IC10	14	3–25	Harford et al. (2011)
			<i>Minutoceillus polymorphus</i>	Growth rate inhibition	72h	21 °C	8.2	NS	IC10	540	46–600	Gillmore et al. (2016)
			<i>Phaeodactylum tricornutum</i>	Growth-rate inhibition	72h	21 °C	8.2	NS	IC10	2100	2000–2200	Gillmore et al. (2016)
			<i>M. polymorphus</i>	Growth rate inhibition	72h	21 °C	8.2	NS	IC10	690	580–800	Golding et al. (2015)
	Chlorophyta	<i>Dunaliella tertiolecta</i>	Growth rate inhibition	72h	21 °C	8.2	NS	IC10	1400	820–1900	Golding et al. (2015)	
	Cnidaria	<i>Acropora tenuis</i>	Fertilization		3h	32 °C	8.2	NS	IC10	2793	1123–4771	Negri et al. (2011)
		<i>Acropora tenuis</i>	Larval metamorphosis		18h	32 °C	8.2	NS	IC10	1263	861–1694	Negri et al. (2011)
<i>Exaiptasia pallida</i>		Reproduction reduction		14 days	28 °C	8.1	6518 ± 15	EC10 ^d	817	440–1480	Trenfield et al. (2017)	

Table 5 continued

Type of organism	Phylum	Tested species	Tested endpoint	Test duration	Temperature	pH	Hardness (mg/L CaCO ₃)	Toxicity measure ^b	Al concentration (µg/L)	Range (µgAl/L)	References
	Arthropoda	<i>Amphibalanus amphitrite</i>	Successful transition of nauplii into cyprid inhibition	96h	28.2–30.8°C	8.2–8.4	NS	EC10	416	368–464	van Dam et al. (2016)
	Mollusca	<i>Saccostrea echinata</i>	Embryo development	48h	29°C	8.2	NS	EC10	410	340–480	Golding et al. (2015)
		<i>Mytilus edulis planiculatus</i>	Embryo development	72h	20°C	8.2	NS	EC10	250	240–260	Golding et al. (2015)
		<i>Nassarius dorsatus</i>	Growth rate	96h	28°C	8.4	6518 ± 15	EC10 ^d	115	91–140	Trenfield et al. (2016)

^aWhere low pH means a pH range between 5.0 and 7.0 and neutral pH means a pH range between 7.0 and 9.0

^bWhere the toxicity measure is the 10% effect concentration (EC10) or the 10% inhibition concentration (IC10)

^cUnfiltered and 3 hours of aging value

^dToxicity of Al at 28°C

NS, Non-specified

(Elangovan et al. 2000; Desouky 2006). For example, Desouky (2006) demonstrated in *L. stagnalis* that the number of granules containing Al is significantly increased upon the exposure of snails to aluminium. These defence mechanisms against metal intoxication were also showed in other mollusks, living in other environments, like marine bivalves (*Argopecten irradians*, *Mercenaria mercenaria*, *Mytilus edulis*, *Pecten maximus*) accumulating intra- and extracellular metallic granules in kidneys (Sullivan et al. 1988), scaphopod (*Dentalium rectius*, Reynolds 1990), or terrestrial snail (*Helix aspersa*) (Brooks and white 1995). In addition, Al embryotoxicity was noted in gastropod *Radix quadrasi* embryos (Factor and de Chavez 2012), which showed growth retardation. Al may also affect the behaviour of freshwater organisms as it was demonstrated for *L. stagnalis* or the bivalve *Anodonta cygnea*, by lowering their physical activity (Truscott et al. 1995; Kádár et al. 2002). Moreover, concerning the crustaceans, the exposure of the freshwater crayfish *Pacifastacus leniusculus* to environmentally relevant concentrations of Al at neutral pH affects its immunocompetence, particularly the ability of the haemocytes to remove bacteria from the circulation (Ward et al. 2006). This disturbance of the crayfish immunocompetence would be mainly due to the previously described hypoxia (Ward et al. 2006). Most of the measured EC₁₀ for freshwater organisms belonging to various phylum (Table 5) is lower than Al environmental concentrations estimated in freshwater ecosystems (Table 1) (Cardwell et al. 2018; Gensemer et al. 2018).

Aluminium effects in marine organisms

In addition to studies carried out on freshwater organisms, others assessed the toxicity of Al on various marine species. Akpiri et al. (2019) established that Al is a genotoxic agent, which induces concentration-dependent oxidative stress to marine sponge cell cultures exposed at least to 0.2 mg total Al/L. In addition, it has been shown to affect reproduction processes of marine organisms, such as sea urchin *Sphaerechinus granularis* in which Al exposure to its sperm induced an inhibition of fertilization success (gametotoxicity) when it is exposed to 10⁻⁴ M of Al (Pagano et al. 1996). This impact of Al on reproduction has also been observed in the cnidarians. Indeed, Trenfield et al. (2017) have

shown that Al induced a reduction in the asexual reproduction of sea anemone *Exaiptasia pallida* with an EC₁₀ of 817 µg dissolved Al/L (Table 2). In addition, Negri et al. (2011) observed that Al induced an inhibition of fertilization (IC₁₀ = 2793 µg/L) and especially of larval metamorphosis (IC₁₀ = 1263 µg/L) in coral *Acropora tenuis*. Moreover, Al is able to induce an embryotoxicity, which has been demonstrated on embryos of barnacles (*Amphibalanus amphitrite*), mussels (*Mytilus edulis*, *Mytilus galloprovincialis*), oysters (*Saccostrea echinata*), and sea urchins (*Paracentrotus lividus*, *Sphaerechinus granularis*) (Pagano et al. 1996; Golding et al. 2015; van Dam et al. 2016). The embryotoxicity was also shown in a marine snail *Nassarius dorsatus*, in which exposure to Al caused a growth inhibition of its larvae with an EC₁₀ of 115 µg dissolved Al/L (Trenfield et al. 2016). For example, van Dam et al. (2016) noted a decrease in the percentage of successful transition of the nauplii into cyprid in the barnacle *Amphibalanus amphitrite* during exposure to increasing concentrations of Al with an EC₁₀ of 416 total µg/L. It was also reported that Al causes growth alteration of various species of microalgae (*Ceratoneis closterium*, *Minutocellus polymorphus*, *Dunaliella tertiolecta*, *Tetraselmis* sp., *Phaeodactylum tricorutum*) (Golding et al. 2015; Gillmore et al. 2016).

Some dissolved Al environmental concentrations (Table 1) already exceed some of the noticed 10% effect (EC₁₀) or inhibition (IC₁₀) concentrations of marine organisms (Table 5). For instance, the EC₁₀ of the marine snail *Nassarius dorsatus* reduced its growth rate (115 µg dissolved Al/L; Table 5) (Trenfield et al. 2016) as well as the IC₁₀ of the marine diatom *Ceratoneis closterium* also concerning its growth rate (14–80 µg/L; Table 5) (Harford et al. 2011; Golding et al. 2015; Gillmore et al. 2016) is below the higher Al environmental concentration measured in coastal waters (131.96 µg dissolved Al/L; Table 1). In that way, these species and perhaps others may already have been affected by the current Al environmental concentrations in marine ecosystems.

Could future climate change affect the bioavailability of aluminium?

Over the past 200 years, human activities such as fossil fuel combustion, cement production, biomass

burning, or agriculture have released anthropogenic carbon dioxide (CO_2) into the atmosphere, increasing the atmospheric CO_2 partial pressure (P_{CO_2}) (Gattuso et al. 1998; Feely et al. 2008). These anthropogenic emissions lead to the uptake of atmospheric CO_2 by oceans, which are one of the sinks for excess atmospheric CO_2 , through air–sea exchange at the sea surface (Gattuso et al. 1998; Provoost et al. 2010; Cummings et al. 2011). As a result, seawater P_{CO_2} increases which causes a shift in the chemistry of seawater and an acidification of the ocean surface (Caldeira and Wickett 2003; Michaelidis et al. 2005; Provoost et al. 2010). In fact, dissolved CO_2 forms carbonic acid (H_2CO_3) by reacting with water molecules, and then H_2CO_3 will dissociate into hydrogen and bicarbonate (HCO_3^-) ions, resulting in a decrease in pH and carbonate ion (CO_3^{2-}) concentration (Gazeau et al. 2011). Because of increasing atmospheric CO_2 concentrations from pre-industrial times, ocean surfaces possibly already have seen their pH decreased by approximately 0.1 Ph unit over the past two centuries (Haugan and Drange 1996; Provoost et al. 2010), leading today to a surface seawater pH ranging from 8.1 to 8.2 (Bates et al. 2014). Furthermore, the evolution of pH at ocean surfaces has also been estimated for the next two centuries: in the year 2300, Caldeira and Wickett (2003) expect a pH reduction of 0.7 units coming from a continuous release of fossil fuel CO_2 into the atmosphere. However, ocean acidification (OA) might not be uniform all over seawater surfaces and may even be accentuated in coastal waters. Indeed, in these environments, in addition to increasing atmospheric CO_2 concentration (Gazeau et al. 2011), the pH decrease in coastal waters can be worsen, for instance, by eutrophication (Cai et al. 2011; Gazeau et al. 2011), the upwelling of CO_2 -rich waters (Feely et al. 2008), the permanent or episodic inputs of acidic river water (Salisbury et al. 2008), and the emission of volcanic vents (Duarte et al. 2013). Therefore, in coastal ecosystems, OA is not exclusively linked to changes in atmospheric CO_2 , which may be a relatively minor component of pH variation (Provoost et al. 2010; Duarte et al. 2013). A decrease in pH in some coastal waters (Tatoosh Island, Dutch coastal waters) has already been observed and shows that the decline of pH could be higher than predicted (Wootton et al. 2008; Provoost et al. 2010). Furthermore, according to Duarte et al. (2013), current models do not adequately

capture the pH dynamics in coastal waters in contrast to the pH of the open ocean that adheres generally to global models (Hofmann et al. 2011).

Aluminium has a highly pH-dependent solubility (Zhou et al. 2018), inducing an increase in Al solubility when pH decreases. Thus, Al becomes more soluble and its toxicity increases (Driscoll and Schecher 1990; Gensemer and Playle 1999; Santore et al. 2017). The acidification of coastal waters could increase Al solubility, concentration, and bioavailability, which could lead Al to become more toxic to marine biota (Gensemer and Playle, 1999). These potential increased effects of Al would probably be added to the negative effects that the acidification can cause. For instance, Massabuau et al. (1987) showed that the acidification of freshwater induced the complete loss of fish population in rivers (region of Cornimont). To our knowledge, few studies have been carried out on the impacts of raised atmospheric CO_2 concentrations on Al toxicity to aquatic animals. For example, Nelson and Campbell (1991) focused on the effects of acidification on the geochemistry of Al, Cd, Pb, and Hg in freshwater ecosystems. It was suggested an increase in Al concentrations caused by acid precipitation, lake acidification, or acid leaching, increasing the solubility of this metal and enhancing its transport from edaphic to aquatic environments (Cronan and Schofield 1979; Driscoll and Schecher 1990; Pyrzynska et al. 1999). Nevertheless, the potential effects of OA to Al bioavailability could exclusively be observed in semi-closed (like the Mediterranean Sea or Bays) or in closed (like lakes) zones where the incidence of pH decrease will be more pronounced. In addition to the effects of OA on pH bioavailability, we can also ask ourselves what would be the effects of the rising temperature caused by global warming. Indeed, an increase in temperature could decrease the rate of the Al^{3+} form, one of the dissolved Al forms, which are bioavailable (Lydersen et al. 1990). Thus, the increase in temperature could change Al speciation in aquatic environments and thus could change its impact on freshwater and marine fauna. For example, Lewis et al. (2012, 2016), Campbell et al. (2014), and Ivanina et al. (2014) showed that ocean acidification increases metal (Cd, Cu) toxicity in bivalves and polychaetes. Lydersen et al. (1990) observed this decrease in the rate of Al^{3+} by increasing the temperature by 25 °C; however, the temperature is predicted to only raise by 2–6 °C on

average (Riebeek 2010), so the change of Al speciation might be minor. Therefore, the impacts of global warming (OA and raising temperature) on Al speciation, bioavailability, and toxicity should be investigated to have a clear view of the evolution of Al toxicity in this context.

Conclusions

Al is present in both marine and freshwater environments in variable concentrations. Under laboratory conditions, the toxic effects of Al on freshwater and marine animals were observed. Indeed, its toxicity affects multiple aspects of animal life (embryogenesis, reproduction, respiration, etc.). Its toxicity could be evolving in response to future ocean acidification; therefore, the study of Al toxicity in the context of ocean acidification should be investigated, especially in the case of semi-closed or close waters (the Mediterranean Sea, lakes, etc.), which could see their pH decrease in a more dramatic way, at least locally.

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Declarations

Conflicts of interest The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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