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Review

Heavy Metal Pollution Monitoring at King Sejong Station, King George Island, Antarctica

In-Young Ahn*, Heeseon J. Choi, and Ko-Woon Kim

Korea Polar Research Institute, KORDI
Ansan P.O. Box 29, Seoul 425-600, Korea

Abstract : The coastal environment of King George Island is potentially subject to contamination by pollutants arising from station operations, such as emissions from fossil fuel burning, oil spills, waste disposal, etc. As a preparatory step to assess such impacts on the marine environment and living organisms of this island, two molluscan species (the bivalve *Laternula elliptica* and the gastropod *Nacella concinna*) were selected as biomonitors for metal pollution monitoring, and their baseline levels have been investigated for the past several years at King Sejong Station. In this review, variability of the baseline levels is discussed in relation to body size, tissue type, and sex. Natural elevations of some metals are also discussed with respect to the environmental characteristics of this region.

Key words : Antarctica, King George Island, *Laternula elliptica*, metal pollution monitoring, *Nacella concinna*

1. Environmental Monitoring in Antarctica

Antarctica is the most pristine environment in the world, but some areas have already been exposed to contaminants of anthropogenic origin (Boutron and Wolffe 1989; Suttie and Wolffe 1993). This is especially the case for King George Island, where eight countries operate year-round stations, and it is expected that there are some sources of heavy metals arising from station operations, such as waste disposal, oil spills, and emissions from fossil fuel combustion. Recently, some signs of heavy metal contamination were detected in station areas on the island. Pb and some other metals were found to have elevated levels in the topsoil and lichen, which was attributed to the combustion of leaded gasoline and the use of spray paint (Hong *et al.* 1999). Signs of metal contamination from local sources have also been detected in the marine environment. Concentrations of Pb, Mn and Mo in marine sediments collected near several stations on this island were found to be highly elevated compared to other Antarctic localities, and the elevated Pb (>70-120 $\mu g\ g^{-1}$ dry weight) was thought to be related to anthropogenic influences (Alam and Sadiq 1993). Lead concentrations were also found to be quite elevated in some inshore waters adjacent to a station with a dramatic decrease towards offshore indicating that the Pb had originated from point sources at the station (Lee *et al.* 1990; KORDI 1998).

Regular and effective monitoring using appropriate biomonitor species is therefore required to assess the impact of metal pollution on the coastal ecosystem of this island and to facilitate early detection of possible unforeseen negative effects on the ecosystem. Article 3 of *Protocol on Environmental Protection to the Antarctic Treaty,* which began to be enforced in 1998, calls for regular and effective monitoring to allow assessment of the impact of on-going activities on the Antarctic environment and associated ecosystems.

2. Biomonitors for Metal Pollution Monitoring in the Antarctic Marine Environment

The first step in metal pollution monitoring would be to elucidate the baseline accumulation pattern of biomonitor species. Mussels and oysters have been used worldwide as

*Corresponding author. E-mail : iahn@kordi.re.kr

biomonitors for pollution in temperate coastal waters under the design of 'Mussel Watch' (Farrington *et al.* 1983; Goldberg *et al.* 1983; Lauenstein *et al.* 1990). These bivalve species, however, are absent in the Antarctic waters. In the Antarctic, the two bivalve species, *Laternula elliptica* and *Adamussium colbecki*, which appear to meet the criteria for the 'Mussel Watch' programs, have been recognized as sentinel organisms for environmental monitoring by virtue of their wide distribution, large body size, and high population density (Mauri *et al.* 1990; Berkman and Nigro 1992; SCAR/COMNAP 1996). The scallop *A. colbecki*, from King George Island has not been reported on however. Only *L. elliptica* is present at the island. Near King Sejong Station, *L. elliptica* is present commonly at depths ranging from 5-30 m, with dense patches at depths of around 20 to 30 m, where sediment consisted of mud mixed with sand and gravel (Ahn 1994; KORDI 2000). This infaunal bivalve species, however, is absent in the intertidal and subtidal zones at depths of less than 5 m apparently due to the frequent impact of ice.

The intertidal environment is probably most vulnerable to the contamination of anthropogenic pollutants from the stations, because all the stations on King George Island are located on the shore. However, little attention has been paid to this area, apparently due to substrate instability caused by ice-related impacts occurring during most of the year and the resultant lack of large-sized life forms. The intertidal environment, however, is an area of ecological importance. The intertidal zone consists mostly of pebbles lacks infauna, but it provides substrates for epilithic or epiphytic diatoms and for filamentous macroalgae (Ahn *et al.* 1997). Benthic diatoms and other microphytobenthos detached and washed from the intertidal pebble substrates may serve as an important food source for shallow water herbivores (Ahn *et al.* 1997). Many tiny invertebrates, such as amphipods and nematodes also inhabit this zone.

The patellid limpet *Nacella concinna* (Strebel 1908) is the most conspicuous and usually the only large invertebrate species found in the ice-impacted and physically unstable intertidal zones. Although not as widely distributed as *L. elliptica* and *A. colbecki* around Antarctica, *N. concinna* occurs commonly in dense patches around the Antarctic Peninsula and adjacent islands (Walker 1972; Picken 1980; Beaumont and Wei 1991; Nolan 1991; Brêthes *et al.* 1994), where station density is higher than anywhere else in Antarctica. *N. concinna* was previously used to monitor the Bahia Paraiso diesel fuel spill in 1989 in Arthur Harbor, where the limpets were found to contain elevated levels of polynuclear aromatic hydrocarbons near the station in the harbor with the highest contaminant levels for intertidal specimens (Kennicutt *et al.* 1992). Najle *et al.* (2000) reported histopathological alterations in the digestive glands of *N. concinna* in an experiment where they were exposed to high concentrations of Cd, and suggested that the histopathological response of *N. concinna* could be used as a biomarker of exposure to metal contaminants. The utility for environmental monitoring studies of other patellid limpets as biomonitors of heavy metal pollution has been reported from other geographic localities (Preston *et al.* 1972; Bryan and Hummerstone 1977; Ramelow 1985; Miramand and Bentley 1992; Campanella *et al.* 2001). In our monitoring studies (Ahn *et al.* 1996, 1999, 2001, 2002) we have chosen the clam *L. elliptica* as a biomonitor for metal pollution monitoring in the shallow subtidal waters, and the limpet *N. concinna* for intertidal areas.

3. Environmental Setting around King Sejong Station

Laternula elliptica and *Nacella concinna* were collected from subtidal and intertidal waters in Maxwell Bay, mostly in Marian Cove. Marian Cove, where King Sejong Station is located, is in northeast Maxwell Bay. The cove is a small fjord-like embayment, which is characterized by a deep U-shaped basin with a maximum water depth of ~100 m. Surface water freezes in winter and melts in summer, but a variable cover of drifting ice occurs during most of the year. Seawater temperatures vary seasonally from a maximum of ca. 1.7°C in February to a minimum of ca. −1.7°C in August; salinity ranges from 32.7 to 35.1‰ (Kim 1996; Kang *et al.* 1997; KORDI 1999).

It should be noted that King George Island is comprised mainly of volcanic rocks enriched with Cu, Mn and Fe, Cu being particularly enriched (>100-186 $\mu g\ g^{-1}$ dry weight) in the rocks near the study area (Jwa and Lee 1992). It has been reported that massive land runoff from the surrounding ice fields occurs every austral summer, influencing considerably the sedimentary processes and physicochemical properties of the nearshore seawater (Chang *et al.* 1990; Ahn 1994; Yoo *et al.* 1999). In summer, the terrestrial volcanic rocks are eroded by the glacial melt water runoff, and lithogenic particles containing Cu and other elements are likely introduced into the bay with the melt-water runoff (Ahn *et al.* 1996). Cu concentrations in the near shore coastal sediment near the study area were highly elevated (68-88 $\mu g\ g^{-1}$ dry weight) and the elevated Cu is believed to originate from the terrestrial volcanic rocks. Further details of the hydrographic features and other environmental conditions of this area have been well described elsewhere (Chang *et al.* 1990; Ahn 1997; Kang *et al.* 1997; Ahn *et al.* 2000).

4. Baseline Concentrations of Heavy Metals in the Biomonitor Species

Effect of body size

Body size can be important in making data comparable, especially when there are considerable differences in metal concentrations among individuals of various body sizes, and therefore should be taken into consideration in sampling and data analysis. *Laternula elliptica* are large in size, growing up to 11 cm in shell length with wet tissue weight up to 80 g. Besides, muscle tissues, most (Fe, Cd, Zn, Pb, Ni, Cr, Co) metal concentrations, which are far below average, constitute the largest fraction ($\approx 50\%$) of the whole tissue mass (Ahn *et al.* 1996, 2000). It would, therefore, be very time-consuming and also not very effective to detect variability among different individuals if whole tissues were used for metal analysis. Therefore, variations of metal concentrations with body size were investigated in kidneys, digestive glands and gills. These three organs are non-reproductive body parts, and were shown to have a strong metal accumulation tendency (Ahn *et al.* 1996).

The concentrations of most metals increased with body size, while an inverse relationship was found only in the digestive glands of Cu, Zn and Mn (Ahn *et al.* 2001). In particular, in the gills, concentrations of Mn, Fe, Cu, Cd, Ni, Cr, Co, and As increased linearly with shell length. Interestingly, as for the kidney, concentrations of Cd, Zn, Pb, Co and Ni were best situated on quadratic regression curves, *viz.* increasing continuously until they peaked at a shell length of around 80-mm (Ahn *et al.* 2001). This may reflect specific regulatory activities for the extremely high accumulation of these metals in *L. elliptica* kidney, particularly Cd (28-354 $\mu g\ g^{-1}$ tissue dry weight, TDW). The extremely high accumulations of Cd in the kidneys implicate that *L. elliptica* should have metal-binding proteins or other intracellular ligands necessary for detoxification.

The overall prevalence of positive relations with body size in *L. elliptica* indicates net accumulation of these metals throughout the lifetime of this species, may be at least in part associated with the slow growth and long lifespan of this cold-water bivalve species. In other bivalves in temperate waters, inverse relationships are more common, which is likely to be the result of the higher metal uptake rates of smaller individuals. The inverse relationships found for Cu, Zn and Mn in the digestive glands of *L. elliptica* also indicate that these metals are associated mostly with particulate matter in the seawater and taken up by feeding.

While positive relations are prevalent in *L. elliptica*, concentrations of the metals in *N. concinna* decreased in general with an increase in body size (Ahn *et al.* 1999). This is likely because the majority (>61-90.5%) of body metals in *N. concinna* are found in its viscera. The highest metal accumulation in visceral organs indicates that most metals are taken up from the diet. In general, smaller organisms display higher feeding activity, and thus end up with higher metal concentrations in proportion to unit body mass. Thus, distinct body size-metal concentration relationships were found in the whole tissue of *N. concinna* and also in the three organs of *L. elliptica,* which in turn suggests that body size should be considered in monitoring studies.

Effect of tissue type

In *Laternula elliptica*, concentrations of most metals (Mn, Fe, Cd, Zn, Pb, Cr, Ni, Co) were highest in the kidneys, followed by digestive glands, gills, gonads and muscles (mostly siphon) (Ahn *et al.* 1996). Concentrations of most metals were higher by one or two orders of magnitude in the kidneys than in any other tissues. Their concentrations in the digestive glands and gills were much lower than in the kidneys, but they were significantly higher than in the remaining tissues. On the other hand, the metal concentrations in the muscle tissues and the gonads were far below the average values for tissue samples as a whole. In general *L. elliptica* was shown to have a strong metal-accumulation tendency.

Significant concentration differences among different body compartments were also found in *N. concinna* with the highest values in the viscera (Ahn *et al.* 2002). Concentrations of Mn, Fe, Cu, Cd, Pb, and Cr in the viscera were greater, mostly by one or two orders, than those in muscles (mostly foot) or in gonads. Metal concentrations in the muscles were similar to those in the gonads except for a few cases. Consequently, the majority of body burdens (>61-90.5%) of Mn, Fe, Cu, Cd, Pb and Cr existed in the viscera, which constituted only 27% of total body mass; this indicates that the metals are taken up mostly in diet. A relatively high burden of Zn (31%) was found in the gonads. Gonads are important sites for Zn accumulation in some marine bivalves (Latouche and Mix

1982) and fish (González *et al.* 1991; Hellou *et al.* 1996; Márquez *et al.* 1998) and the Zn concentrated in the gonads likely perform some functions during gonad maturation or prespawning phases. Latouche and Mix (1981) considered Zn as a key element in the reproductive cycle of *Mytilus edulis*, concluding that Zn was transferred from somatic to gonadal tissue to perform some biochemical reactions during the reproductive season. Thus, seasonal variation of tissue distribution of Zn and other metals associated with reproductive cycles has been reported in bivalves (Latouche and Mix 1981; Martinčič *et al.* 1987; Páez-Osuna *et al.* 1995; Frías-Espericueta *et al.* 1999) and other patellid limpets in temperate waters (Miramand and Bently 1992). In our studies, *N. concinna* were collected in summer prior to spawning when the gonads of both sexes of *N. concinna* were extremely ripe, and the major spawning was yet to come. Tissue concentrations of Zn and other metals in *N. concinna* would, therefore, be expected to change after spawning. A seasonal study may clarify this.

Effect of sex

Variations in baseline metal concentrations according to sex were investigated only for the the limpet *Nacella concinna*. *L. elliptica* was not considered in the study in terms of sex effects, as it is hermaphroditic. In tissue as a whole, the female limpets showed higher concentrations for Zn, Cu, Mn, Pb and Cr, while no element was found to be higher in the males (Ahn *et al.* 2002). Metal analysis for the various tissues showed that the differences between the sexes were largely due to the differences in their gonads. In particular, the differences in Zn, Cu and Mn between the female and the male were attributable exclusively to the differences in the metals in their gonads. Significant concentration differences between the sexes, with higher values in the female, were frequently found in marine molluscs in the case of biologically essential elements such as Cu, Zn, Fe and Mn (Watling and Watling 1976; Orren et al. 1980; Latouche and Mix 1982). As with this study, these differences between the sexes seem to exist mostly in the gonads during reproductive seasons. In the present study, the gonads of both sexes were extremely ripe and were likely at a spawning stage, and the differences in these elements in the gonads were probably greater than at any other time of year. A seasonal study associated with its reproductive cycle may however be necessary to clarify the differences between the sexes.

Concentration differences by sex in *N. concinna*, however, are not very large, because the majority of the metal burden existed in the viscera, which constituted only about 27% of the total body mass. Even in the case of Mn, concentrations were 6 times higher in the female gonads than in the male gonads, while concentrations in the tissue as a whole differed only by 20%. Dissecting out the gonads will eliminate the concentration differences found in the sexes, but this would be cumbersome and very time-consuming. Instead of removing gonads, Latouche and Mix (1982) suggested, from their study on the mussel *Mytilus edulis*, that when differences due to sex are not too significant, the differences can be minimized if the male to female ratio within the sampled population is kept constant.

5. Elevation of Some Metals in the Biomonitors of King George Island

Both the bivalve *Laternula elliptica* and the gastropod *Nacella concinna* from King George Island were shown to strongly accumulate most heavy metals analysed (Mn, Fe, Cu, Cd, Pb, Zn) with tissue concentrations being comparable to those of their related species in temperate waters (Ahn *et al.* 1996, 1999). In spite of signs of lead contamination in the near shore surface water, as previously reported (Lee *et al.* 1990), we didn't find any conclusive evidence of lead contamination in the two biomonitor species. On the other hand, we found that some metals, in particular Cd and Cu concentrations, were highly elevated in these two Antarctic mollusks as compared to those of their related species in temperate waters.

Cadmium

The Cd concentrations in the tissues of *L. elliptica* and *N. concinna* were found to be highly elevated in comparison with those found in their related species (Ahn *et al.* 1996, 1999, 2001). The Cd concentrations throughout the tissue samples were found to have ca. 7.5 $\mu g\ g^{-1}$ tissue dry weight in *L. elliptica* (Ahn *et al.* 1996) and 10.7 $\mu g\ g^{-1}$, TDW in *N. concinna* (Ahn *et al.* 1999). The elevated Cd levels found in our studies, however, seem natural rather than anthropogenic. Similar values were reported for *N. concinna* collected from various sites on King George Island (Moreno *et al.* 1997). Elevated Cd levels were also reported in the Antarctic scallop *Adamussium colbecki* (26-49 $\mu g\ g^{-1}$, TDW) in Terranova Bay in the East Antarctica (Mauri *et al.* 1990). Elevated Cd levels were reportedly a unique feature of Antarctic marine herbivorous organisms (Honda *et al.* 1987; Mauri *et al.* 1990; Berkman and Nigro 1992; Ahn *et al.* 1996; Bargagli *et al.* 1996;

Moreno *et al.* 1997) living in naturally Cd-enriched waters. The Cd concentrations in the Southern Ocean, which are several times higher than in any other oceans, can be accounted for by the upwelling of nutrient-enriched deep water to the surface (Orren and Monteiro 1985; Honda *et al.* 1987; Fowler 1990). Cadmium, which has a high affinity to phosphate (Orren and Monteiro 1985), seems to be taken up by phytoplankton along with phosphate, and consequently, accumulates in herbivores grazing on the primary producers and is biomagnified through the food chain. Cd levels comparable to those in contaminated areas were also reported in Antarctic macroalgae and other invertebrates in the Antarctic Peninsula region (Moreno *et al.* 1997). Cd accumulation in the tissues of *L. elliptica* would, therefore, be expected to vary with the extreme seasonal cycle of primary production. In addition, Antarctic animals like *L. elliptica* and *N. concinna* are known to grow very slowly compared to their related species in temperate waters. For a same-sized individual, these Antarctic species are likely to be older than the related species in temperate waters, and have probably accumulate metals over a longer period of time. This may contribute in part to the elevation of Cd levels in *L. elliptica* and *N. concinna*.

The Cd concentrations in *L. elliptica* kidneys (28-354 $\mu g\ g^{-1}$ TDW in Ahn *et al.* 2001) are extremely high, being comparable to those reported from the kidneys or the digestive glands of other bivalves exposed to unnaturally high Cd concentrations in experimental media (Scholz 1980; Köhler and Riisgård 1982; Nolan and Duke 1983; Langston and Zhou 1987; Viarengo *et al.* 1987). Although much lower than in the kidneys, Cd concentrations in the digestive glands (12.6-28 $\mu g\ g^{-1}$ TDW) and in the gills (0.88-5.4 $\mu g\ g^{-1}$ TDW) of *L. elliptica* are also relatively high, given that the study area has been least affected by human activities. Mollusks accumulate metals principally in two organs: the digestive glands and kidneys. The extremely high accumulation of Cd indicates that *L. elliptica* should have metal-binding proteins or other intracellular ligands necessary for detoxification (Roesijadi 1992, 1996). Metallothioneins or other cytosolic ligands have been identified, following exposure to cadmium in several tissues, particularly the kidneys, digestive glands, and gills of bivalves (Scholz 1980; Köhler and Riisgård 1982; Nolan and Duke 1983; Viarengo *et al.* 1987; Roesijadi and Klerks 1989) and of gastropods (Langston and Zhou 1987; Bebianno *et al.* 1992). In the case of the Antarctic scallop *Adamussium colbecki*, Cd accumulations were mostly associated with particulate fraction and cytosolic metallothionein in the digestive glands (Viarengo *et al.* 1993) and existed in typical lysosome-like bodies in the kidneys (Nigro *et al.* 1992). Interspecific variations would be expected in view of metal-binding ligand systems, and future studies are needed to elucidate the nature of metal-detoxificating ligands in *L. elliptica*, exposed to the naturally Cd-enriched environment.

Cu and other metals from lithogenic sources

The Cu levels have been found to be elevated in both *L. elliptica* and *N. concinna* from our studies (Ahn *et al.* 2001, 2002) and also from *N. concinna* of other sites on King George Island (Moreno *et al.* 1997). This has been attributed to a regional feature occurring during austral summer, *viz. inflow of melt-water laden with Cu and other metals originating from the Cu-enriched lithogenic sources* (Ahn *et al.* 1996). Berkman *et al.* (1992) also reported that trace metal concentrations were higher in the shells of the Antarctic scallop *Adamussium colbecki* found in shallow water adjacent to glacial meltwater runoff, suggesting that the significant decreases in the concentrations of Fe, Mn, Cu and Zn at certain depths could be caused by the decreasing influence of the glacial runoff offshore.

Cu was highly elevated particularly in the muscles and digestive glands of *L. elliptica* and in the viscera of *N. concinna* (Ahn *et al.* 1996, 2001, 2002). Processes or mechanisms related to Cu accumulation in the muscles of *L. elliptica* are to be resolved. On the other hand, the Cu elevation in the visceral organs of *L. elliptica* and *N. concinna* suggests that Cu accumulation occurs mainly as a result of food or other particulate matter in seawater being ingested. In strong support of this, a recent seawater analysis in the study area showed that the majority of Cu in seawater in summer was associated with particulate matter, most of which were lithogenic particles introduced into the bay with glacial melt-water runoff from terrestrial volcanic rocks (KORDI 2001). Both the *L. elliptica* and *N. concinna* used in our studies were collected during the mid-summer period when the melt-water inflow is likely to be at its maximum for the year and feeding activity is apparently stronger than in any other season (Ahn 1993, 1997). The magnitude of melt-water influence on the tissue accumulation of Cu and other metals derived from lithogenic sources would, however, be expected to vary with time or season. Thus, the background levels of Cu would be expected to be highly variable (Martincic *et al.* 1987). Seasonal factors should, therefore, be taken into consideration in elucidating the baseline accumulation pattern of these metals in the biomonitor species.

6. Summaries

1) Metal concentrations in the two biomonitor species vary highly according to size, body part, and sex, which should be taken into consideration in establishing a monitoring program. In the case of *L. elliptica*, tissue compartments particularly gills, digestive glands, and kidneys would be better biomonitors than would whole tissue samples.

2) Cd is naturally elevated in these two herbivorous biomonitor species as with other Antarctic herbivorous organisms. In particular, the kidneys of *L. elliptica* accumulate extremely high concentrations of Cd. Accumulaion and detoxification of Cd in *L. elliptica* kidneys is a subject worthy of future study.

3) Cu and other metals of lithogenic sources are also found to be elevated in these two biomonitor species from the King George Island, which can be attributed to the local geochemical features, *metal-laden ice-melt water runoff during austral summer.* The amount and composition of the melt-water runoff, however, would be expected to vary with time and space, causing spatial and temporal variation in the tissue levels of these metals in the biomonitor organisms. A seasonal study would, therefore, be needed to elucidate their baseline accumulation patterns.

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