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Article *in* Marine Pollution Bulletin · August 1996

DOI: 10.1016/0025-326X(96)00026-4

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PII: S0025-326X(96)00026-4

Marine Pollution Bulletin, Vol. 32, Nos 8/9, pp. 631–635, 1996

0025-326X/96 $15.00+0.00

Seasonal Variations in the Concentrations of Cu, Cd, Pb and Zn in *Arctica islandica* L. (Mollusca: Bivalvia) from Kiel Bay, Western Baltic Sea

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Seasonal variations in the concentrations of Cu, Cd, Pb and Zn in soft dry tissues of the ocean quahog, *Arctica islandica*, from Kiel Bay, Germany were studied. Significant monthly variations in the concentrations of the four metals were observed. The two essential metals, Cu and Zn, had their maximum values during the summer months, when the dry soft tissue weight of *A. islandica* was reaching its maximum values as well. The highest values of the non-essential metals, Cd and Pb, were observed in winter when the dry soft tissue weight of *A. islandica* was reaching its minimum values. Copyright © 1996 Elsevier Science Ltd

Since the beginning of metal pollution studies in the aquatic environment, inshore organisms have been considered to be subjected directly to pollution and thus have been intensively studied. Offshore organisms, on the other hand, have been considered to be living in a clean environment away from sources of pollution and thus have remained poorly studied. Recently, it was found that some offshore organisms from the central North Sea have higher concentrations of some metals than those collected from the nearshore areas (Claussen, 1987; Kroencke, 1987; Borchardt, 1988). Rainbow & Phillips (1993) concluded in their recent review about cosmopolitan biomonitors that mussels and oysters of the genera *Mytilus*, *Perna* and *Crassostrea* are among the best-studied cosmopolitan biomonitors. They emphasised the need for considering other species and taxa for use in regional, national or international monitoring programmes. This motivated developing offshore biomonitors to monitor trace metal concentrations in the offshore areas.

The importance of the ocean quahog, *Arctica islandica* in the western Baltic has been documented in different studies (Arntz & Weber, 1970; Arntz, 1973, 1977, 1980; Brey *et al.*, 1990). Its use as a potential biomonitor in the western Baltic was first introduced by Swaileh & Adelung (1994). The baseline levels of Cu, Cd, Pb and Zn from different stations in Kiel Bay and the effect of body size on the concentrations of these metals were provided. The present study continues the last-mentioned work by studying the seasonal variations in the concentrations of Cu, Cd, Pb and Zn in soft tissues of *A. islandica*. This will make understanding and explaining results of future monitoring programmes based on this species easier.

Materials and Methods

Sampling protocol and analytical procedure

A total of 604 ocean quahogs, *A. islandica*, (shell length range 30–60 mm) were caught by dredging on a monthly basis from Kiel Bay at water depths ranging between 21 and 23 m. Sampling was carried out over a 1-year period that started in July 1992 and ended in June 1993. Quahogs were cleaned of mud, frozen and transported to the laboratory. There, specimens were thawed before shell length and dry weight of each specimen were determined. Shell length was measured as the greatest anterio–posterior distance measured to the nearest 0.1 mm. Dry weight was determined as weight of the whole soft tissues determined to the nearest 0.1 mg after 1 week freeze-drying and 24 h oven-drying at 60°C. For metal analysis, sub-samples (nearly 20 mg) of the dry soft tissues of *A. islandica* were weighed and digested in TEFLON tubes with TEFLON covers that allow acid reflux to take place. Digestion took place in a block heater. The digestion mixture used was composed of nitric:perchloric super pure acids (Merck; ratio: 1:1, v:v). The volume of the mixture used was always 10 times the sample's weight. Temperature was increased gradually up to 200°C over the first 5 h. Subsequently, digestion was allowed to continue for a further 10 h. Finally, samples were dried via evaporation and the volumes were brought to 1 ml using 1 N HCl. Solutions were filled in polyethylene Eppendorf reaction vessels and kept in a refrigerator at 4°C for later analysis. At the time of metal measurement, subsequent dilutions using 0.2% HNO_3 were performed. Then the concentrations of the four metals, Cu, Cd, Pb and Zn, were measured with a flameless atomic absorption spectrophotometer type Perkin–Elmer 3030 with automatic sampler and Zeeman background corrector. *Mytilus edulis* tissue, CRM 278, provided by the Commission of the European Communities, Community Bureau of References, Brussels and Copepod Homogenate, MA-A-1 (Tm) no. 645, provided by the International Atomic Energy Agency, International Laboratory of Marine Radioactivity, Monaco were used as reference material. All the accepted recoveries of the four metals were above 90%.

Statistical treatment of the data

All weight measurements refer to the dry weight of soft tissues. Statistics were performed using the SYSTAT-computer program (Willkinson, 1992).

Results and Discussion

To be sure that a possible seasonality is not due to variations in mean shell size from month to month, analysis of variance (ANOVA) was applied. Monthly samples did not show any significant difference in their mean shell lengths (Fig. 1A). The monthly mean shell length ranged between 42.9 (July) and 47.1 mm (September). Next, variations in dry soft body weight were tested where significant monthly variations (ANOVA, $p < 0.0001$) were observed (Fig. 1B). Quahogs collected in September had significantly higher mean dry weight than those captured in November, February, March, April and May, whereas all other pairwise comparisons between months were not significantly different (Tukey test, $p < 0.05$). An increase in the dry body weight of *A. islandica* first became noticeable in late spring (May) and continued until the maximum mean weight (0.707 g) was reached in September. Thereafter, a drastic decrease in the dry body weight began, which extended until November. Subsequently, a small increase in dry weight again occurred until January before mean dry weight dropped and the minimum mean weight (0.374 g) was reached in March. The increase in dry body weight from May to September (Fig. 1B) coincided with the period of gonadal development of this species (Jones, 1981; Mann, 1982; Rowell *et al.*, 1990), food availability and an increase in bottom water temperature. Some authors found that *A. islandica* started spending their gametes when the bottom water temperature reached approximately 13.5°C (Loosanoff, 1953; Jones, 1981). In the present study, it is possible that *A. islandica* started spending their gametes massively, thus loosing weight, after September (Fig. 1B), when the bottom water temperature measured at 21 m water depth reached nearly 13°C (U. Fiedler, IfM, pers. comm.). The drastic weight loss started in September and continued until November. Thereafter, a slight increase in weight occurred, which might indicate that some food is still available, before another period of weight loss started in January and continued until March. This period of weight loss could be due to food shortage.

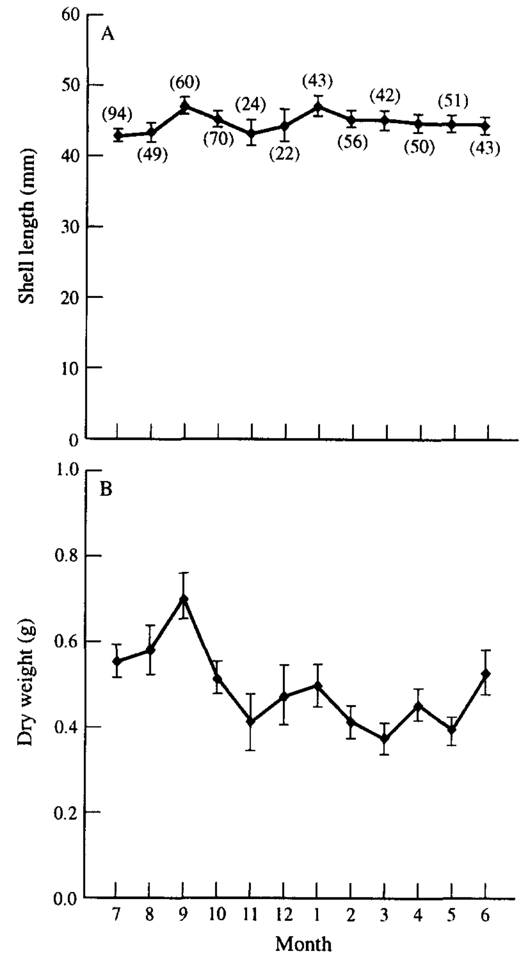

Fig. 1 Monthly variations in (A) mean shell length (mm) and (B) dry soft body weight (g) of *Arctica islandica* samples collected from Kiel Bay and analysed for their metal concentrations from July, 1992 to June, 1993. Number of specimens analysed are shown in brackets.

Recently, Fritz (1991) studied variations from month to month of dry soft body weight of *A. islandica* off New Jersey. He observed no clear seasonal trend in any condition index calculated for standard-sized individuals of 95 mm length. But generally, his specimens collected in spring and summer had higher visceral weight, that included the gonad, than those collected in autumn and winter. He found that this difference in weight originated mostly from gonadal development but not from somatic tissue growth.

Significant seasonality (ANOVA, $p < 0.0001$) in the concentrations and contents (body burden) of the four metals in soft tissues of *A. islandica* was observed (Fig. 2). Both concentrations and contents followed similar trends. Contents, however, seem to vary less than concentrations with time of the year.

Cu concentrations in *A. islandica* started increasing after May, reaching the maximum value (18.6 $\mu g\ g^{-1}$) in August (Fig. 2A). Thereafter, a drastic decrease in the concentration of Cu occurred until the minimum value

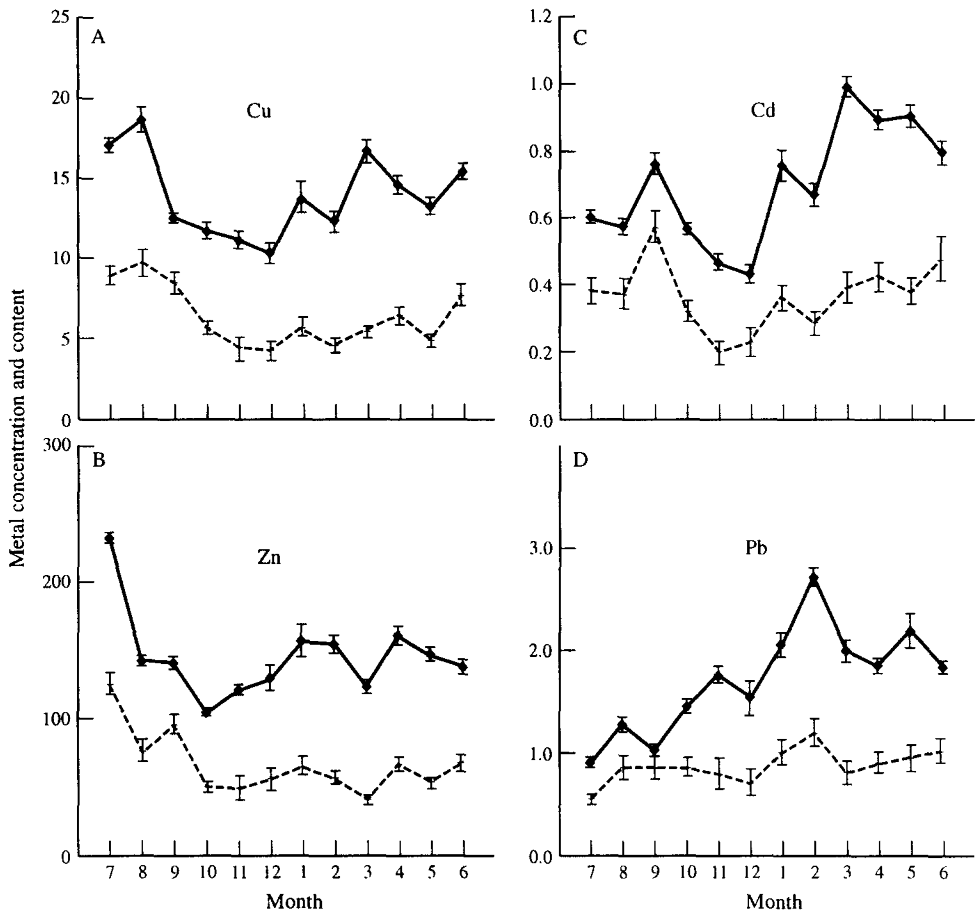

Fig. 2 Monthly profiles for the concentrations in μg g^{-1} (solid lines) and contents in μg (broken lines) of Cu, Cd, Pb and Zn in *Arctica islandica* samples (shell length 30–60 mm) collected from Kiel Bay from July, 1992 to June, 1993. For number of specimens analysed each month see Fig. 1A.

(10.1 μg g^{-1}) was reached in December. Subsequently, Cu concentrations increased again to reach a new maximum in March. The ratio between the maximum and the minimum mean concentration is 1.8. The seasonal variation profile for Zn looks similar to that of Cu (Fig. 2B). The maximum mean concentration (231.8 μg g^{-1}) was observed in July and the the minimum one (103.6 μg g^{-1}) occurred in October. Again, a slight increase in Zn concentrations was observed during the winter months. The ratio between the maximum and minimum mean concentration is 2.2.

Concentrations of these two essential metals, Cu and Zn, had two periods of increase, one main period in summer and another smaller one in winter. The first period of increase in metal concentration corresponds to the increase in dry weight of *A. islandica* indicating that these two metals are accumulated when new tissues are assimilated and might be associated with gametogenesis. This is supported by losses in the concentrations and contents of Cu and Zn at the time of spawning, which indicates loss of these metals with gamete spending. The other period of increase in the concentrations of these metals, which was observed in winter, could be caused by the loss of weight of *A. islandica* during the food shortage period in winter.

High Cd concentrations were observed in September, decreasing through December. Concentrations increased towards the end of winter. The highest Cd mean concentration (0.992 μg g^{-1}) was observed in March, concentrations remaining high until June. The lowest mean metal concentration (0.429 μg g^{-1}) occurred in December. The ratio between the highest mean concentration and lowest one is 2.3. Generally, high mean Pb concentrations occurred in winter and early spring and low mean concentrations occurred in summer and autumn. The highest monthly mean concentration (2.7 μg g^{-1}) occurred in February while the lowest monthly mean (0.91 μg g^{-1}) was observed in July. The ratio between these two is 3.

The increase in body weight of *A. islandica* in late spring and summer (Fig. 1B) seems to dilute the concentrations of the non-essential metals, Cd and Pb, since their concentrations in winter and early spring, when dry body weight was minimum, were clearly higher than those in summer and autumn (Fig. 2C, D). Similar results were obtained by different authors (Boyden & Phillips, 1981; Cain & Luoma, 1990). It is important to note that at the time of spawning, and thus weight loss, a drastic decrease in the concentrations and contents of Cd occurred, while Pb concentrations and contents remained unaffected. This might be explained as metal loss with the gametes at spawning (Boyden & Phillips, 1981).

Seasonal variations in the concentration of metals in molluscs is well-known (Bryan, 1973; Frazier, 1975, 1976; Phillips, 1976, 1977, 1980; Orren *et al.*, 1980;

633; Goldberg *et al.*, 1983; Cain & Luoma, 1986, 1990). Phillips (1980) mentioned three inter-related factors that cause seasonality of metals in the biota. These are pollutant delivery (run-off), organism physiology, particularly sexual cycle and changes in ambient water quality like temperature and salinity. Bryan (1973) studied the seasonality of trace metals in scallops. He found the highest concentrations in autumn and winter and suggested that metal concentrations were inversely related to phytoplankton productivity. Fowler & Oregioni (1976) studied trace metals in *Mytilus galloprovincialis* and found the maximum concentrations in spring. They attributed this to the reproductive state of the organisms and to the high winter run-off. They found the ratio between seasonal maximum and minimum concentrations to be greatest for Cr (factor of 8.8) and minimum for Zn (factor of 2). Other studies (Phillips, 1976, 1977; Denton & Burdon-Jones, 1981) concentrated on temperature and salinity as causative agents for seasonality in metal concentrations. Strong seasonal variation in metal concentrations in *Macoma balthica* was found to be associated with seasonal changes in soft tissue weight (Cain & Luoma, 1990). Similarly, seasonal variation in metal concentration in the oyster *Crassostrea gigas* found to be mainly due to changes in the weight of the whole soft body parts, which in turn were due to gonadal development and spawning (Boyden & Phillips, 1981). Maximum metal concentrations in *M. edulis* from the North Sea occurred in late winter and minimum concentrations in autumn. The range of variation is nearly 2–3 fold (Borchardt *et al.*, 1988).

In the present study, it is not likely that run-off could have affected metal concentrations in the water and as a result in *A. islandica* since the stations are located in the offshore area. Seasonality in metal concentrations could be due to changes in the water quality, temperature and salinity, or most probably due to variations in body weight of *A. islandica*, which coincided with the reproductive cycle and food availability. Reproduction of molluscs is accompanied by variations in the biochemical composition (lipids, carbohydrates and proteins; Gabbott, 1983), which cause changes in the affinity of these compounds to metals (Oesterberg, 1974). In *A. islandica*, the ratios between means, maximum and minimum concentrations of the metals ranged between 1.8 (for Cu) and 3 (for Pb). This is in good agreement with ratios calculated for mussels in general and found to be between 2 and 3 (Borchardt *et al.*, 1988).

This research was carried out as a part of a Ph.D. Thesis, Institute for Marine Sciences at the University of Kiel, Germany. The author would like to thank. Prof. Dr D. Adelung for his supervision and valuable comments. The financial support of the German Academic Exchange Service (DAAD) is highly acknowledged. Thanks are due to the personnel of the research ship *Littorina*, Institut für Meereskunde, Kiel, who were always helpful and co-operative in obtaining the samples.

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