

Salinity effects on cadmium concentrations in blue mussels in the Baltic Sea

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Photo: Benutzer Dakone, 2003

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Abstract

This bachelor's essay is a review written at the Swedish University of Agricultural Sciences on suggestion from the Swedish Museum of Natural History.

The purpose of this study was to increase the understanding of how the salinity in the Baltic Sea affects the concentrations of cadmium (Cd) observed in blue mussels, *Mytilus edulis*, and to be able to predict more accurate future trends of the bioavailable Cd. Cadmium is a heavy metal that deposits to the Baltic Sea by runoff, point sources and atmospheric deposition. Cadmium is toxic to aquatic organisms and may bioaccumulate.

Blue mussels feed by filtering the water. Through the filtration the mussel is contaminated by chemical pollutants, which means that chemical analysis of mussels can reflect how contaminated a habitat is. The mussels are suitable biomonitors since they are sedentary, easy to collect, abundant and large enough for tissue analysis.

In the Baltic Sea there is a salinity gradient with decreasing salinity from west to east and from south to north. The surface water has lower salinity compared to the deep water. The mussel size is affected by the salinity with smaller mussels in less salty waters due to stress.

The amount of soluble cadmium in water is increasing with salinity since the salt is competing with Cd for adsorption sites on particles and the formation of soluble chloride complexes are increasing. However, since the chloride complexes are not bioavailable, the bioavailability of cadmium is increasing with decreasing salinity.

At the surface zooplanktons take up Cd from the water and in deeper layers it is released again by the decomposition by detritus, creating a vertical gradient. In deeper waters, below the detritus, the concentration is relatively constant.

While using organisms to examine pollutants, biological variations must be considered. One way to eliminate influences from individual differences is to put the cadmium concentration in the soft tissue in relation to the mussel shells. Different body parts contain different concentrations of Cd. How the concentration is divided between the body parts is however indefinitely, since different experiments have shown different results.

Keywords: Cadmium, Cd, salinity, blue mussels, *Mytilus edulis*, the Baltic Sea.

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1 Introduction

1.1 Purpose

In this study the salinity's effect on the concentration of cadmium in blue mussels (*Mytilus edulis*) in the Baltic Sea (Figure 1) was examined. The Baltic Sea is a very sensitive ecosystem due to the brackish water with its high salinity gradients and the fact that some of the water can remain in the Baltic for more than 30 years. The increasing concentrations of heavy metals in the Baltic Sea are a threat to the environment. Cadmium (Cd), among other toxic heavy metals released in the Baltic Sea, remains there for a long time and may bioaccumulate in organisms (Westerbom *et al.*, 2002; HELCOM, 2007). The purpose of this study is to increase the understanding of how the salinity of brackish water affects the concentrations of Cd measured in blue mussels and to be able to predict more accurate future trends of bioavailable cadmium.



Figure 1. The Baltic Sea drainage area. (Map: Hugo Ahlenius, UNEP/GRID-Aredal)

http://www.grida.no/graphicslib/detail/baltic-sea-drainage-basin_aacb

1.2 Cadmium

Cadmium is a heavy metal spread naturally by volcanic activity and weathering of cadmium-bearing rocks. Cadmium is also spread anthropogenic by for instance metal production, combustion of fossil fuels and during poor management of waste (Bignert *et al.*, 2014). In the industry the metal is used, among other things, as stabilizer in plastics, in color pigments, as an anticorrosive agent, at the production of nickel-cadmium batteries and during the manufacturing of phosphate fertilizers where phosphate rocks containing Cd are used (Godt *et al.*, 2006; Mar & Okazaki, 2012). Riverine runoff, point sources and atmospheric depositions are the main supplies of Cd to the Baltic Sea. Cadmium is toxic for aquatic organisms and may bioaccumulate. It is one of the metals that has to be analyzed according to both the OSPAR- and the HELCOM-conventions (HELCOM, 2007; Bignert *et al.*, 2014).

1.3 Blue mussels

The blue mussels have existed in the Baltic Sea for about 7000 years. Today the mussels represent 80% of the body weight of all the invertebrates in the Baltic Sea. Despite this the mussels are smaller compared to the mussels living in the Atlantic. The size of the blue mussels in the Baltic Sea varies with salinity, and they cannot live in salinities below 4.5‰. Due to the low salinity mussels do not grow more than up to 3 cm, meanwhile a mussel in the Atlantic may be 10-15 cm (Figure 2) (Westerbom *et al.*, 2002; Tedengren, 2008).



Figure 2. Three blue mussel specimens, from Skagerrak (left), the Kattegat (middle) (estimated ages 4-6 years) and the central Baltic Proper (estimated age 12-14 years). (Photo: Qianfen Xiao)

The males and females release their germ cells in the water during the summer and the larva are swimming around for a couple of weeks. This gives the larvae an effective spreading before they settle on hard surfaces down to 30 m depth.

Blue mussels feed by creating a water stream with their gills and then filter the water. One single mussel can filter several liters of water every hour and together the mussels in the Baltic can filter a volume equal to the inland sea at least once a year (Tedengren, 2008). Through this filtration the mussels are provided with phytoplankton, bacteria, algae, parasite larvae and particles, but also chemical pollutants as toxins. This means that chemical analysis of mussels can reflect how contaminated a habitat is. The mussels are one of the most investigated organisms within the marine world due to some beneficial qualities. The mussels are sedentary which makes them representative for a certain environment and easy to collect. They are abundant, have a shell which makes them hard enough to handle in experiments and are long lived (which may also be a problem, see 3.4). They are also large enough for tissue analysis (Luoma & Rainbow, 2008; Brenner *et al.*, 2014).

2 Method

This bachelor's essay is a review written at the Swedish University of Agricultural Sciences on suggestion from the Swedish Museum of Natural History. The information used in the review has been collected from scientific articles, reports, books and websites. Both national and international sources have been used.

3 Results

3.1 Cadmium in the Baltic Sea

The cadmium that enters the Baltic Sea remains there for a long time and the concentrations can be about 20 times higher compared to the North Atlantic. The total Cd load was 41 tonnes in 2004, of which 86% were waterborne and 14% airborne (Figure 3 & 4). The waterborne loads include point sources directly into the sea and loads from rivers. The river loads are measured at the river mouth and includes both upstream point sources like industries, and diffuse sources like agriculture, managed forestry and natural background sources. The airborne loads do not only include cadmium from the catchment area of the Baltic Sea (Figure 1), but also from sources outside the area (HELCOM, 2007). Other estimates presents that the waterborne input of cadmium was 47,7 tonnes in 2006 (HELCOM, 2011), and the airborne input in the Baltic Sea catchment area was 6 tonnes (1 tonne in the Baltic area) (Andersson *et al.*, 2012).

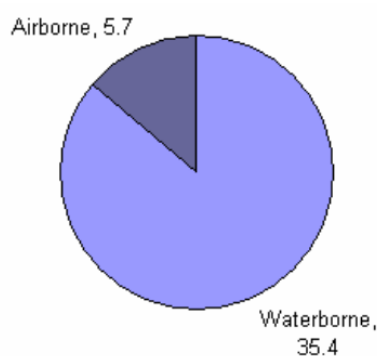


Figure 3. The total Cd load to the Baltic Sea in tonnes during 2004. The values from Russia, Latvia and Lithuania used in the calculations were based on a five year average for the waterborne loads (HELCOM, 2007).

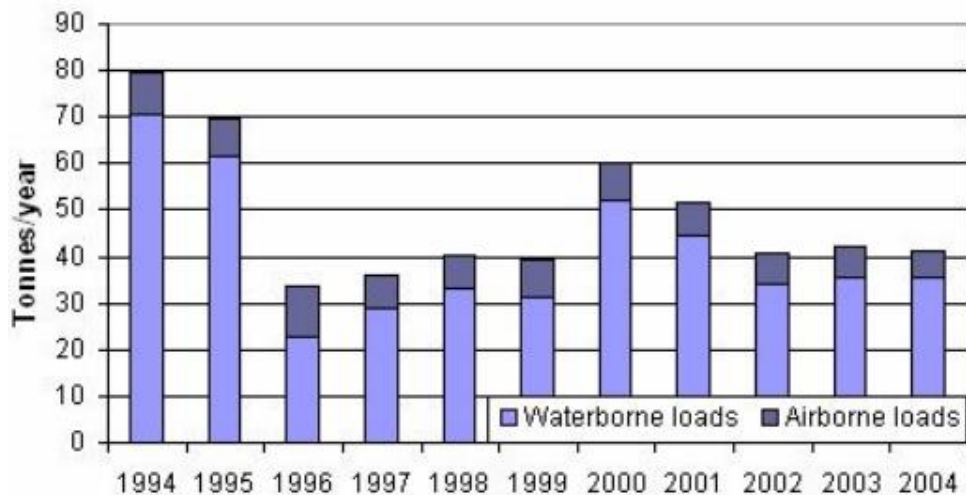


Figure 4. Total waterborne and airborne loads of Cd to the Baltic Sea between 1994 and 2004. In 2000 improved monitoring techniques were used for the waterborne loads (HELCOM, 2007).

3.2 Cadmium in the aquatic environment

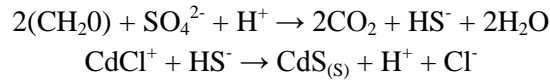
When pollutants are spread in aquatic systems there are interactions between hydraulic, chemical soluble-solid and biological factors. Figure 5 is demonstrating a representation of the cadmium cycle. Cadmium may occur in high concentrations close to harbors and industrial areas. In harbor sediments concentrations of more than 100 ppm have been measured (Manahan, 2010)

In water cadmium can exist as hydrated ions ($\text{Cd}^{2+} \cdot 6\text{H}_2\text{O}$), bound in colloids (i.e. are captured in filter with the size range 0.10-0.45 μm), with humic acids as organic complexes or as inorganic complexes (Parkman *et al.*, 1998; Luoma & Rainbow, 2008). The Cd^{2+} ion's speciation can be affected by factors as pH, the hardness of the water, redox potential, suspended particulates and organic matter (Nriagu & Sprague, 1987).

In freshwater, Cd forms complexes with CO_3^{2-} and OH^- , but with increasing salinity cadmium exists almost only with chloride and also as CdCO_3 complex. Only a minor fraction exists as the free Cd-ion (Nriagu & Sprague, 1987; Stumm & Morgan, 1996). Cd is a very mobile metal and 90% of the total Cd in the Baltic Proper exists in dissolved phase (i.e. can pass through a 0.45 μm filter) (Parkman *et al.*, 1998; Luoma & Rainbow, 2008).

Cadmium is adsorbed to organic matter in oxygen-rich water and form oxide or hydroxide complexes (Bignert *et al.*, 2014). The temperature and salinity differs between surface and bottom water and the difference is increasing during the summer which makes the water more stagnant. That, in combination with decom-

posing of phytoplankton and algae, causes lower oxygen content in the bottom water. The low oxygen concentration leads to a microbial reduction of sulfate of organic matter (CH₂O) which creates sulfides. This results in a precipitation of insoluble cadmium sulfide (Manahan, 2010; Moksnes *et al.*, 2013; Raymond *et al.*, 2013):



The cadmium is bioavailable as the free Cd²⁺-ion. When the salinity is increasing the formation of chloride complexes will increase. The salt ions are also competing with cadmium for adsorption sites on suspended particles. These two factors result in that the concentration of soluble Cd is increasing when the salinity is increasing. The chloride complexes are however not bioavailable, which means that increasing salinity will decrease the amount of bioavailable cadmium (Bignert *et al.*, 2014). The concentration of the free Cd ion is varying in the Baltic Sea due to the variation of the salinity. In the northern Baltic Proper the free ions can constitute 10-15% of the total cadmium, meanwhile in the Bothnian Sea the concentration is 50% and in the Bothnian Bay 70-95% (Parkman *et al.*, 1998).

Cd can be potentially available in sediments and suspended particulate matter such as Cd in inorganic solid phases (e.g. CdCO₃ and Cd(OH)₂), Cd precipitated or co-precipitated with hydrous oxides of manganese and iron, and Cd chelated and insoluble organic bound. It can become mobilized and bioavailable if the conditions are changing, for instance changes in pH, redox potential, microbial action or by erosion, dredging or bioturbation (Nriagu & Sprague, 1987).

Particles play an important role in the transportation of metals because of their high surface area where the metals bind to functional surface groups (Stumm & Morgan, 1996).

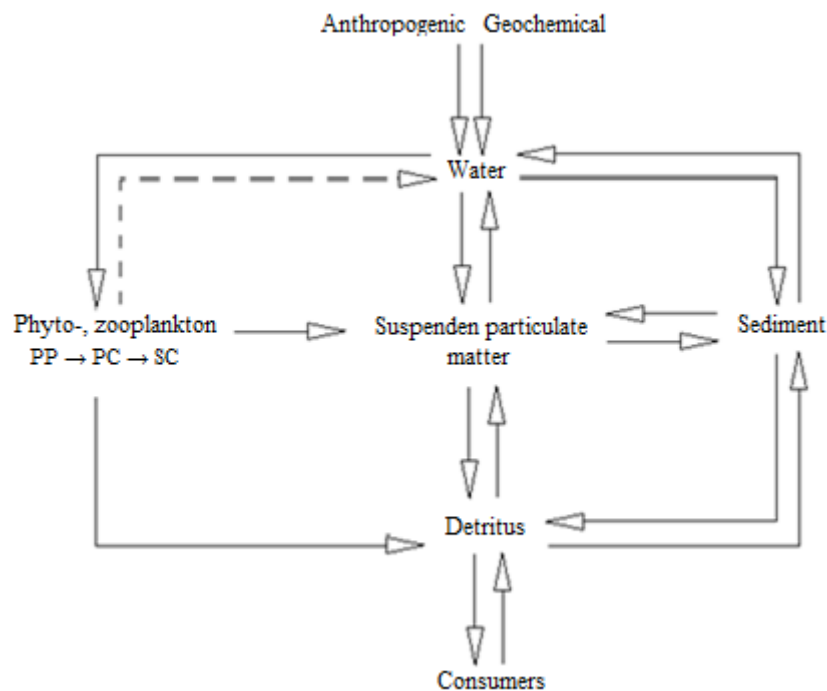


Figure 5. Cadmium cycle in marine environment, PP = primary producer, PC = primary consumer, SC = secondary consumer. After: (Nriagu & Sprague, 1987) Figure 1, chapter 9)

The concentration of cadmium also varies vertically in oceans and is affected by biological activity. A correlation with phosphate is affecting the Cd concentrations when the metal is cycled through organisms. Detritus can incorporate and release Cd in proportion to the regeneration of nitrate and phosphate. The amount of dissolved cadmium in the surface layers is affected by adsorption onto particles and/or uptake by phytoplankton. Zooplanktons are consuming the phytoplankton and store the Cd alternatively release it with faecal pellets. The concentration of dissolved Cd is increasing with depth when sinking detrital particles undergo decomposition and the Cd is once again released. Due to this decomposition, the cadmium concentration is increasing a lot in the blue mussel's habitat since they live down to 30 m depth, where the decomposition is strong. Below the detritus, the concentrations are decreasing slightly and remain at relatively constant values. The primary producers at the surface receive cadmium and nutrients by atmospheric deposition, but also by transport from the detritus regeneration by the vertical mixing and advection of the water. How great the vertical concentrations differ varies between different areas. The concentration gradient is affected by physical oceanographic conditions, how intense the mixing is and the presence or absence of vertical convection. Deep waters and sediments receive cadmium from particles

that settle below the thermocline without complete regeneration (Nriagu & Sprague, 1987; Luoma & Rainbow, 2008).

3.3 The salinity of the Baltic Sea and its effects on mussel size

The salinity of the Baltic Sea is regulated by the inflow of salty water from the Danish Straits and freshwater inflow from mainly the northern and eastern parts of the Baltic. This results in a salinity gradient with decreasing salinity from west to east and from south to north. There are also temporal differences in salinity, both seen over the year and between different years as the inflows are irregularly combined with meteorological factors. The mussel size is also changing with the salinity. The blue mussels are becoming smaller from west to east. In the Gulf of Finland (Figure 6) more than 98% of the population was 2-10 mm (median length: 3 mm, quartile Q_1 : 2mm, Q_3 : 4 mm). In the Archipelago Sea the mussels were bigger (median length: 8 mm, Q_1 : 4 mm, Q_3 : 13 mm). The size difference between the Gulf of Finland and the Archipelago Sea was abrupt. Mussels collected at Tvärminne had a median length at 3 mm (Q_1 : 3 mm, Q_3 : 4 mm) while mussels only 20 km away collected at Hangö västra had a median length at 6 mm (Q_1 : 4 mm, Q_3 : 10 mm). This can be compared to Söderskär, 130 km from Tvärminne, where the mussels were only 1 mm smaller with a median length at 2 mm (Q_1 : 2 mm, Q_3 : 3 mm). The size difference is continuing along the salinity gradient. At Utö the median length was 9 mm (Q_1 : 5 mm, Q_3 : 15 mm) (Westerbom *et al.*, 2002).

The surface water has a lower salinity compared to the deep water. In the Baltic Proper the surface water has remained at a salinity of about 7 ‰ since 1990, meanwhile it has been a slight decrease in the Gulf of Bothnia. In the deep water in the Bornholm Basin the salinity has decreased since a peak in 2003 while it has remained high in south eastern Baltic Proper, eastern Gotland and northern Baltic Proper. The difference between the surface water and the deep water has increased since the 1990's, which holds back vertical mixing (Figure 7) (Andersson, 2014).



Figure 6. Map of the Baltic Sea with locations where mussel size was measured. (Figure: Relief Map of Baltic Sea by Nzeemin, NordNordWest. Modifications of map: Linda Johansson.)

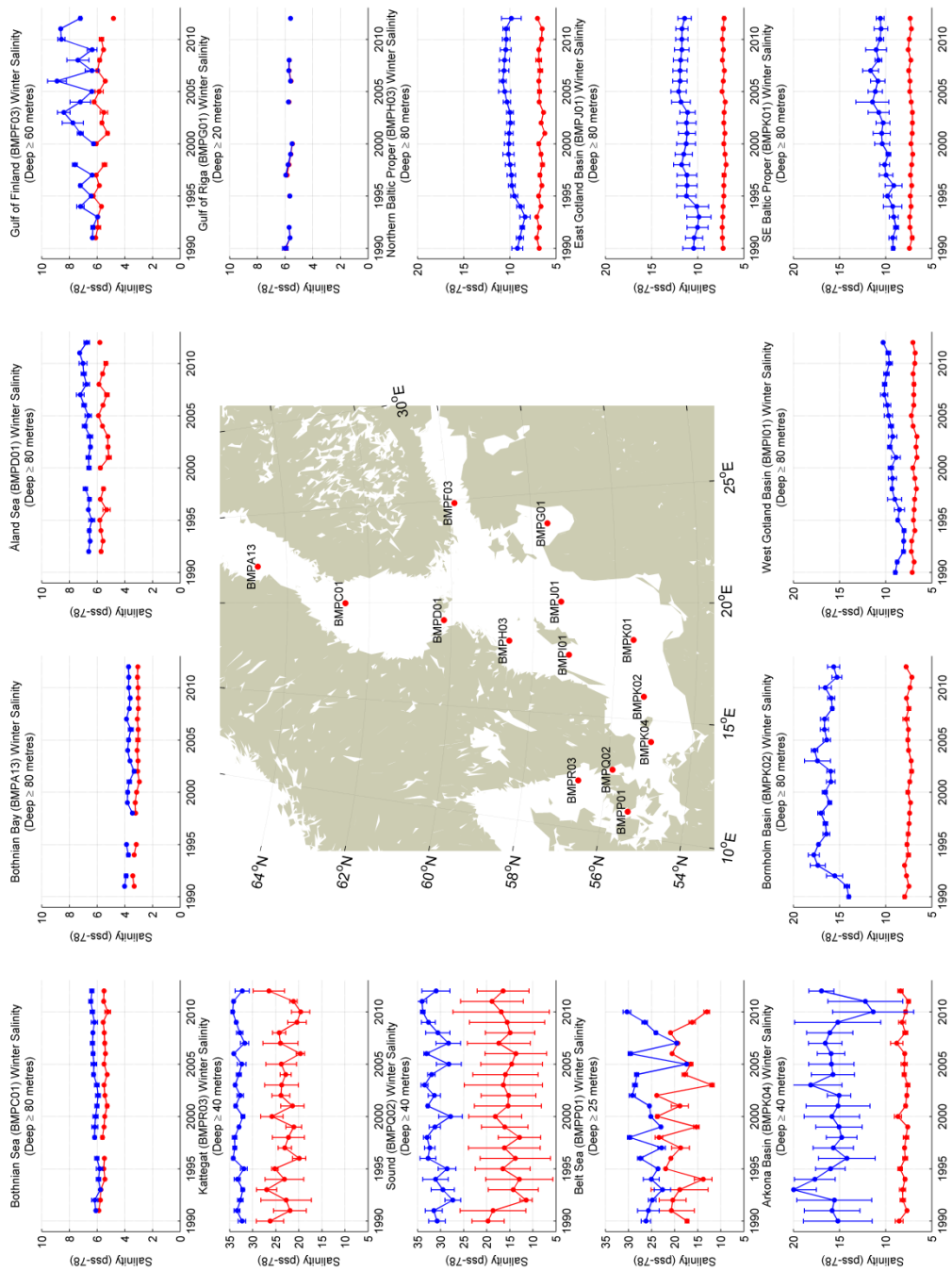


Figure 7. Temporal variations of the salinity in the Baltic Sea, 1990-2012. Red = surface <math>< 10\text{ m}</math>, blue = deep-water. (Andersson, 2014)

3.4 Bioavailability and mussels as bioindicators

In this review the definition of bioavailability is “a relative measure of the total ambient metal that an organism actually takes up when encountering or processing environmental media, summated across all possible sources of metal, including water and food as appropriate” (Luoma & Rainbow, 2008).

Most of both the internal and the external surfaces of mussels consist of epithelium. The cadmium uptake is determined by the amount of the metal that is absorbed by the cell, i.e. passes the cell membrane of either the body surface or the gut epithelium. If the metal only attaches to the cell, is adsorbed, it does not play any role in ecotoxicity (Luoma & Rainbow, 2008).

To be able to pass the cell membrane does the dissolved metal needs to be transported through membrane proteins: either carriers or transporters, since the membrane itself is impenetrable to dissolved metals. This transport is called a passive facilitated diffusion, and is due to a concentration gradient with lower concentration inside the cell. The gradient is of a specific chemical form of the metal and not by the total amount of the metal. Inside the cell there are many proteins and the metal is bound to them, which maintains the concentration gradient. The higher concentration differences the higher the uptake (Luoma & Rainbow, 2008).

To be able to measure how great the uptake of metals by food is the assimilation efficiency (AE) is determined. The AE varies between mussels depending on how much food they get through the filtration, how contaminated the food is and how much of the metal that is extracted and assimilated into them. The mussel’s choice of food and how contaminated it is are also affecting factors, as how the ingestion rate and digestive strategy is (Luoma & Rainbow, 2008).

The definition of bioaccumulation is the “net accumulation of a chemical into the tissues of an organism as a result of uptake from all environmental sources, i.e. both food and ambient water” (Luoma & Rainbow, 2008). This means that the bioaccumulation is the net difference between all forms of uptake and loss integrated over time. The bioaccumulation is a good integrative measure of the degree of exposure to metals by organisms, since the metals do not break down to simpler forms. The amount of trace metals that are accumulated differs between different organisms, but all aquatic invertebrates are accumulating trace metals (Luoma & Rainbow, 2008).

The methods and instruments used to detect metal concentration have developed throughout the years. Almost all data from before the late 1970s are questionable.

During 1950s and 1960s methods and instruments to detect low concentrations of trace metals were developed. In the 1970s laboratories interested in environmental chemistry installed atomic absorption spectrophotometers, but the samples got contaminated which resulted bad quality of the data. The first contamination-free measurements were made in 1976-1977. After that the methods spread and more accurate measurements could be made (Luoma & Rainbow, 2008).

While using organisms to examine pollutants biological variations must be considered. When an organism is changing in weight the metal concentration is affected as well (Figure 8) (Luoma & Rainbow, 2008).

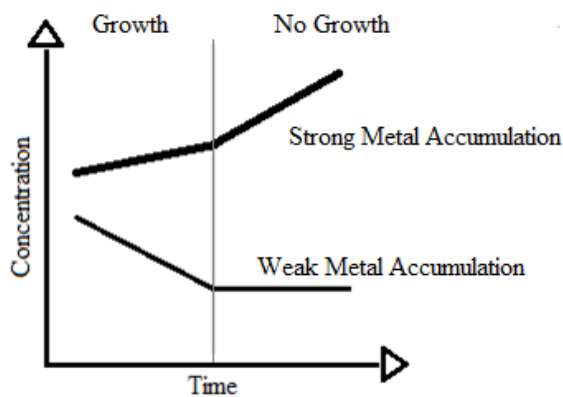


Figure 8. Changes in metal concentration with strong respectively weak accumulation and the influence of, or absence from growth. (After: Luoma & Rainbow, 2008 Fig. 7.1)

One way to eliminate influence from individual differences in size, access to food, spawning and tidal exposure is to put the cadmium concentration in the soft tissue in relation to the mussel shells, the so called Cd/shell-weight index. The salinity has also an effect on calcification, which must be considered (Fischer, 1986). A study by Fischer (1986) showed that when the salinity was below 15 ‰ the increment of shell weight was reduced.

The concentration of Cd in the mussels is influenced by factors as shell length, dry weight percentage, soft body fat percentage, salinity and age (Figure 9). The age can be determined by the layers in the shell, but it is more difficult to determine the age of the blue mussel compared to other bivalves (Xiao *et al.*, 2012).



Figure 9. Some of many factors affecting the mussels and the cadmium concentration.

The benefits of using organisms as indicators are that they show the concentration of bioavailable cadmium and also reflect the condition during a longer period of time (Figure 10). Cadmium concentrations determined from seawater, sediment and suspended matter is a complex matter and does not provide a realistic picture, as the concentrations vary when the physical conditions and biological activities are changing (Figure 11) (Nriagu & Sprague, 1987; Askman *et al.*, 2003).

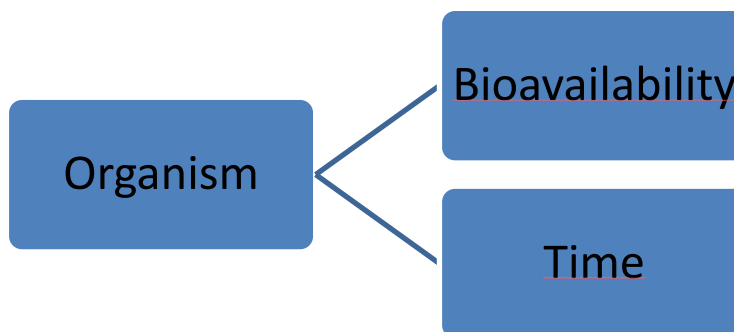


Figure 10. The information provided from doing measurements of heavy metals in organisms.

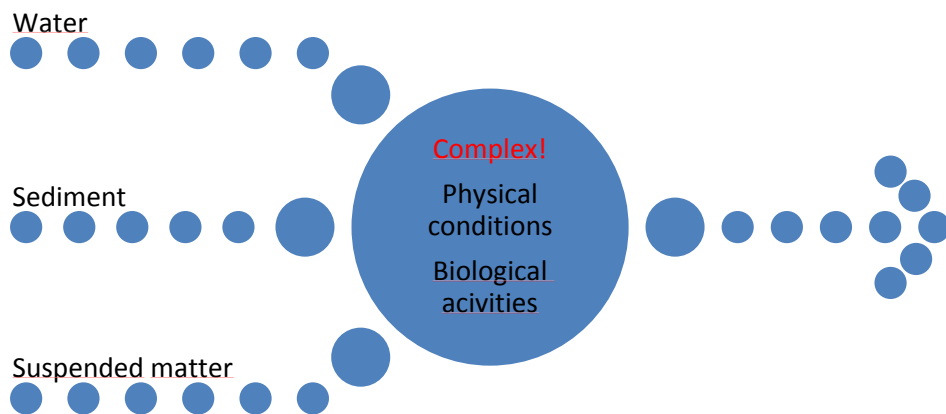


Figure 11. The disadvantages of not using an organism while doing measurements of heavy metals.

Nriagu and Sprague (1987) present different studies of the division of the Cd concentration in the mussel body. One measurement showed that the concentration of Cd in bivalve species is divided as gills > viscera > muscle. The gills are however not showing the true uptake even if it is the primary site of uptake. The Cd content of the gills also shows the adsorption to mucus sheaths, which is being renewed constantly. To the viscera comes material that has passed through the gills. Most of the cadmium in the viscera can be found in the digestive gland and in the kidneys. Another study reported the order kidney >> viscera > gills >> mantle > muscle and a third reported midgut > gill > kidney > mantle > foot > adductor muscle. These two last concerned *M. edulis* specifically (Nriagu & Sprague, 1987).

An organism can avoid a certain kind of food if it is highly contaminated by metals. The high concentration can inhibit digestive enzymes, reduce the feeding rate or negatively affect the transport of food through the digestive tract. These factors can lead to misinterpretation of both the dietary bioaccumulation and toxicity tests (Luoma & Rainbow, 2008).

3.5 The significance of cadmium concentration

A concentration factor (CF) can be used to express the optimum efficiency of accumulation in an organism. In a study (George & Coombs, 1977) the CF in mussels was estimated by using radioactive cadmium chloride. The CF was calculated by dividing the radioactivity in mussels with the radioactivity in water (cpm Cd/g dry weight divided by cpm Cd/g sea water). In the study mussels from the

estuary of the River Ythan, Scotland, were exposed to low and high concentrations of cadmium. At low concentrations the uptake had a linear correlation between time and Cd-concentration. When the CF was above 0.7 µg/ml Cd in sea water there was a decrease in uptake. The decrease indicates that the binding capacity is saturated. At that point visible signs of toxicity started to show too, as froth (George & Coombs, 1977). The decrease in uptake could also be explained by acute toxicity.

The free cadmium ion is the driving force of the bioavailability, but other metals are also present in the water, competing with the Cd. This means that the bioavailability also is controlled by relative ion activities (Nriagu & Sprague, 1987).

3.6 Mussels from different habitats acclimatized to other salinities

In an experiment (Blackmore & Wang, 2003) Cd uptake from both the dissolved phase and the particulate phase was investigated in green mussels, *Perna viridis*, from Hong Kong coastal waters. In the study mussels from two habitats, one with high salinity (28 and 32‰) and one with low salinity (6 and 8‰), had been acclimatized to four different salinities: 10, 17, 23 and 30‰. The study showed that increased salinity lead to decreased Cd influx from the dissolved phase, but also that mussels from habitats with low salinity had a lower influx of Cd compared to mussels from high salinity habitats after acclimatization to the same salinity when the salinity was 17-30‰. There was no significant difference between the two habitats after acclimatization to salinity of 10‰.

The experiment also included clearance rate. The clearance rates was measured by putting the mussels in cadmium-free water and then measure the cell density at different times to see how much cadmium the mussel got rid of. The clearance rate was higher in the mussels acclimatized to higher salinity. The mussels from the low salinity habitat had a higher clearance rate compared to the mussels from high salinity.

Concerning assimilation from ingested diatoms no significant difference between the two populations were shown in the experiment. Mussels kept close to their natural salinity did though have a higher assimilation efficiency compared to mussels acclimatized to a new salinity.

4 Conclusions

The amount of soluble cadmium in water is increasing with salinity since the salt is competing with Cd for adsorption sites on particles and the formation of soluble chloride complexes are increasing. Since the chloride complexes are not bioavailable, the bioavailability of cadmium is decreasing with increasing salinity. In freshwater cadmium also forms potentially bioavailable complexes in the sediment and suspended particulate matter (Nriagu & Sprague, 1987; Parkman *et al.*, 1998; Luoma & Rainbow, 2008).

The concentration of Cd vertically in the ocean depends on biological activities. At the surface zooplanktons take up Cd from the water and in deeper layers it is released again by the decomposition of detritus, creating a gradient. In deeper waters, below the detritus, the concentration is decreasing to relatively constant values. The cadmium can be transported back to the surface by vertical convections and mixing of the water (Nriagu & Sprague, 1987; Luoma & Rainbow, 2008).

Organisms can accumulate metals from both food and ambient water. The bioavailable amount of metals may pass through the cell membrane and into the cell. In invertebrates this can happen through permeable body surface or through the diet via the alimentary canal (Luoma & Rainbow, 2008).

In the Baltic Sea there is a salinity gradient with decreasing salinity from west to east and from south to north. The mussel size is affected by the salinity, with smaller mussels in less salty waters (Westerbom *et al.*, 2002).

While using organisms to examine pollutants biological variations must be considered. Factors as shell length, age, dry weight percentage, soft body fat percentage and salinity can influence the concentration of Cd in the mussels. One way to

eliminate influence from individual differences in size, access to food, spawning and tidal exposure is to put the cadmium concentration in the soft tissue in relation to the mussel shells, a so called Cd/shell-weight index (Fischer, 1986; Luoma & Rainbow, 2008).

Different body parts contain different concentrations of Cd. How the concentration is divided between the body parts is however indefinite, since different experiments have shown different results (Nriagu & Sprague, 1987). Further investigations in the area would be useful.

The low salinity in the Baltic Sea results in stress for animals that are not adapted to such environment. The organisms are in addition exposed to the toxic cadmium (Nriagu & Sprague, 1987). An interesting area to investigate is what happens when focus is not only on Cd, but Cd in combination with other metals as well. Will the toxic effects change?

More research could also be useful in inter-population differences in metal uptake, uptake from particulate phase and the salinity's effect of metal efflux from mussels.

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