

ECOTOXICOLOGY OF STABLE POLLUTANTS IN AFRICAN MARINE ECOSYSTEMS

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Summary

Molluscs (particularly bivalves) and fish are usually proposed as good bioindicators for environmental contamination of marine ecosystems. Different steps can be recognized when assessing such contamination. The first approach consists of comparing stable residue concentrations determined in various environments or different periods. In this chapter, this is illustrated for heavy metals in bivalves from Africa. For some of them, species differences could be detected making comparison difficult: such data can only be used for comparing orders of magnitude. More detailed discussions need the data to be first corrected (normalized) as a function of the main factor influencing accumulation, which means that the transfer, excretion and accumulation mechanisms have first to be identified and understood. Such factors vary, however, with the type of residues, mainly in their liposolubility, and also with the biota concerned: age, lipid content, sex, weight, etc. The importance of this step, as well as the difficulty of coming to clear conclusions, are illustrated for heavy metals, with emphasis on the non-essential ones—mainly mercury—in bivalves. The special case of mercury contamination of the sardine in the Mediterranean Sea is also presented. Finally, the importance of speciation and detoxification as well as the "biomass effect" of phytoplankton are discussed.

From the practical point of view, i.e. in comparison with legal norms as generally accepted, the situation can be considered as generally safe for African fish and molluscs. Important exceptions should, however, be mentioned among the heavy

metals: they concern mainly West Africa, with high levels of zinc, mercury and cadmium in bivalves from the Ivory Coast, and cadmium in most countries for which data are available: Ivory Coast, Cameroon, Ghana, Nigeria and South Africa. Even if mean concentrations are not often above the legal limits (norms), they are already of the same order of magnitude and are thus worrying. Moreover, some age classes are expected to be above the norm and should thus not be suitable for human consumption. The same can be said about mercury in sardines from the Mediterranean coast of North Africa: even if the situation is less acute than in the northern part of the sea, the largest individuals accumulate Hg at concentrations close to the norm and might pose a problem. Finally, local problems of Hg in fish and bivalves can be expected in regions of important manual gold digging, mainly in rivers, but possibly in surrounding marine systems as well.

Massive and preventive use of organochlorine pesticides (DDTs) probably represents the main pollution stress in developing countries, including the African ones. This concerns both the direct consequences of any non-specific pesticide, namely the potentiality to reduce biodiversity by destroying non-target organisms, and the indirect ones mainly due to bioaccumulation, and leading possibly to sterilization and/or feminization of the higher trophic levels.

1. Introduction

Two main types of large scale and long-lasting pollution threats can be recognized at the global level: on the one hand, organic pollution leading to high organic content in aquatic ecosystems and, in the long term, to eutrophication, i.e. an increase of nutrient concentration, and as a consequence of primary production, oxygen depletion. These effects are obvious in zones and cities with high human density, if insufficient water treatment plants are in service. In an ecotoxicological approach, this chapter is more devoted to the second type of pollution, namely the bioaccumulation of stable residues such as organochlorines—pesticides and PCBs—and heavy metals in African marine ecosystems.

Whereas in western countries, most organochlorine pesticides were banned from agriculture in the early 1970s, they are still exported to and massively used in Africa. Although some of them have a more than doubtful reputation where it concerns environmental and human health, they are still reasonably cheap tools to control crop damage by pest organisms or to control vectors of infectious diseases such as onchocerciasis and malaria. They were basically replaced by organophosphate pesticides in western countries. Their continuing utilization in developing countries is to be understood by different factors, starting with the very bad impression that they are considered too dangerous for rich countries, but not for developing ones... Other aspects are, however, that the utilization of organophosphates, which unlike organochlorines, have strong direct toxicity, must be preceded by a rigorous programme of training for the users (the farmers) in order to avoid serious accidents. Finally, some acute environmental problems exist in Africa, such as malaria, and this may explain the application of DDT, while actually the situation in western countries was never that acute. But alternative solutions, based for instance on biological control, deserve more effort and investment, potentially leading to limitation of pesticide use—they are still

used massively and preventatively, i.e. including when they are not actually necessary. One should also take into account that the main effect of organochlorines and organophosphates is probably a decrease of biodiversity, due to their lack of specificity: they do not "just" destroy the pest insects one tries to get rid of, but many other species as well. In the long term, this might in fact become the main environmental threat of pesticides.

Due to their very high ecological stability (the actual half-life of organochlorine pesticides ranges from a few decades to a century), and a high affinity for the lipid fraction of organisms, organochlorine pesticides continue to accumulate in aquatic ecosystems, years or decades after the end of their use on land. Direct effects on human health when not applied with the necessary precautions (as for other pesticides) include neurotoxic and respiratory effects; long-term (life-time) effects are disruption of hormonal function as well as induction of cancer (teratogenic and carcinogenic effects).

A similar group of man-made organochlorine chemicals, the polychlorobiphenyls (PCBs), are known to behave in a similar way once released into the environment. For decades, industrial electric transformers contained a mixture of oil and PCBs at extremely high concentrations, up to 33%, for their high thermostability and isolation properties. In order to degrade PCBs without production of toxic byproducts, special ovens must be used, with temperature strictly controlled above 1600 °C. One of the byproducts of burning at relative low, uncontrolled temperatures are dioxins, probably the most harmful and persistent organic pollutant. Major accidents with oils with high PCB content in developing countries could be traced back to the re-use of these oils (tapping from old industrial transformers) and the subsequent burning of it. In the author's opinion, this is also the probable explanation for the 1999 major "dioxin crisis" in Belgium.

Two major groups are to be recognized, and treated separately, among the heavy metals. The essential ones are necessary to any biological compartment (for example to enzymes and other proteins), at low concentrations (e.g. for bivalves: 1 to 2 mg/ l of Zn, and 0.05 to 0.2 mg/ l of Cu. At higher concentrations, however, they become toxic. The most obvious example is probably the necessary presence of iron, Fe, in the hemoglobin of our blood.

Although of natural origin, non-essential heavy metals, particularly mercury, cadmium, and lead, (Hg, Cd and Pb respectively) are known for their high toxic effects, even at low exposure rates. Given the fact that they are used in a large series of applications, especially Hg and Cd, some of the material inevitably ends up in the environment and the human food chain. Given the low level of industrialization in Africa, the risk of large scale heavy metal pollution is expected to be low.

Only mercury, which is used in small scale gold digging activities throughout African countries (Tanzania, Ghana, etc.) might pose a direct threat to aquatic—mainly freshwater—ecosystems and subsequently to human health through the consumption of fish and molluscs. Mercury salts are added to gold-containing clay in order to extract and bind the gold. Heating of this amalgam separates both with loss of mercury fumes. The risks are numerous: direct exposure through skin contact and inhalation of large

doses of inorganic Hg and Hg fumes leading to neuro-intoxication and interfering with cellular respiration. Impact on river ecosystems can be severe, after direct and indirect losses and accumulation in river sediment, with possible biomethylation and subsequent accumulation effects through the aquatic food webs (freshwater, and in the final stage, marine ecosystems). The questions remain as to whether this use of Hg may be the cause of the dramatic decline of several common fish species (direct toxicity or behavioral effects and indirect effects through prey shortage), and what is the effect on human health of consumption of mercury-contaminated fish. On the other hand, the Mediterranean Sea is known for its high Hg content, even if it is often attributed to natural sources: this is why the Hg content in North African regions deserves more careful attention.

Ecotoxicological studies of stable residues ideally involve investigation of large scale toxicological problems, at the ecosystem level. Since the study of a complete ecosystem is basically impossible, they mostly make use of bioindicators, i.e. biological material the contamination of which is supposed to directly reflect the wider environmental contamination. Moreover, fishery products tend to accumulate some heavy metals like Hg at high levels, making them the main source of contamination for humans, even if local conditions also influence human Hg contamination, such as volcanic activities (including geothermy), gold digging or the amount of dental fillings using Hg amalgam. This is why fishes are often proposed, as well as molluscs, in order to evaluate both the environmental pollution, and possible threats to human health. Molluscs also offer the advantage of being sessile and thus to reflect the contamination of a limited geographic zone, while many fish species have seasonal migrations and other movements, during which they can be exposed to regions with different levels of contamination.

It is not possible to give a comprehensive report on such a broad topic, and on the other hand, it is not satisfactory to present general ideas or conclusions, without showing the basic data on which they are built. This is why some aspects will be presented in some detail as examples. There will be a progressive development of the discussion and interpretation of data on heavy metals in bivalves and fish, in order to illustrate the successive steps of the discussion.

2. Organochlorines

2.1. Pesticides

Pesticide concentrations were determined in a bivalve from Nigeria: the African bloody cockle *Anadara (Senilia) senilis* (Azokwu and Joiris, unpublished data). Most concentrations were very low, around the detection limit of the method ("traces"): α HCH, γ HCH (lindane), δ HCH, aldrin, dieldrin, endrin, heptachlor epoxide. Heptachlor was not detected, which might reflect the absence of significant recent utilization.

As expected, the most abundant pesticide was DDT, as well as its metabolites: p-p'DDT (the form which is used as insecticide), o-p'DDT, p-p'DDE, o-p'DDE, o-p'DDD and p-p'DDD. For the whole group (_DDT), values up to 0.3 μ g/g dry weight (dw) i.e. 0.3 "ppm" were noted. Since DDT is slowly metabolized into the more stable form DDE, the DDE to _DDT ratio provides information on the residence time of these residues in

the environment: the ratio of 0.3 found in this case was very low, reflecting as expected recent DDT applications. This value can be compared with the 0.6 ratio registered in bivalves from Latin America (Sericano *et al*, 1995).

Such data are to be considered as preliminary, and should thus not be discussed in detail. The order of magnitude, however, appears to be high: during a large scale "mussel watch" in Central and South America, oysters collected at only 10 stations, out of a total of 120, had DDT values higher than 0.1 $\mu\text{g/g dw}$, with a maximum of 0.2. Orders of magnitude could also be compared with results of a "mussel watch" in the heavily polluted North Sea, where we detected maximal concentrations of 0.03 $\mu\text{g/g dw}$ for DDT, and 0.1 for DDE (this team: unpublished data).

For a more detailed discussion of the data, one must take the basic transfer and accumulation mechanisms into account, such as age (total body length), sex, condition index and lipid content. It is only after such a normalization of the results that further comparisons become possible, like geographical, seasonal and temporal variations. This will be illustrated later for heavy metals (see further).

2.2. PCBs

During the same study (Azokwu and Joiris, unpub.), PCB concentrations were also determined, with maximal concentrations of 0.45 $\mu\text{g/g dw}$ when expressed as the sum of 9 congeners, corresponding to 1.3 $\mu\text{g/g dw}$ if calculated as "total" PCBs, i.e. as an Aroclor 1254 standard mixture.

For comparison, the data collected during the same "mussel watch" in Central and South America, showed that only 15 stations out of 120 showed PCB concentration in oyster higher than 0.1, and two were above 1.0 (Sericano *et al*, 1995). In the North Sea, we registered total PCBs concentrations between 0.1 and 0.5 $\mu\text{g/g dw}$ (this team: unpublished data). This shows again that African marine systems are heavily contaminated not only by pesticides, but also by PCBs.

3. Heavy metals

[Preliminary remark: the discussion of heavy metal data should mainly concern the non-essential ones such as Cd and Hg, and to a lesser extent Pb, since they normally do not play any physiological role, and their presence in significant amounts is thus a reflection of anthropogenic pollution (with the possible exception of Hg in the Mediterranean Sea). For methodological reasons, however, Cd and Pb are often determined together with the essential heavy metals, and Hg separately. This is why, respecting the origin of the results, such data will be presented with the other metals, and Hg separately.]

As an example, bivalve contamination by heavy metals will be shown, and the different steps of the discussion developed, from very broad and general to more detailed. A general overview of heavy metal concentrations in African molluscs is presented in Table 1.

Species	Origin	Cu	Zn	Fe	Mn	Pb	Cd	Reference
Oysters	Morocco					0.07	0.07	Chafik <i>et al.</i> , 1998
	Ghana	3.1	460	80			0.17	Biney, 1991
	Ghana	4.4	175	60	1.8		0.06	This team (2)
	Nigeria	5.8	630			0.2	0.25	Okoye, 1991
	Cameroon	8.5	410		3.0		0.25	Mbome, 1988
	Ivory Coast	24	1200				0.65	Metongo, 1991
	South Africa	2.4	210			0.1	0.21	Watling and Watling, 1982
Cockles	Morocco					0.7	0.04	Chafik <i>et al.</i> , 1998
	Ghana	1.0	13	11	1.6		0.19	Biney, 1991
	Ghana	0.9	7.3	110	2.0		0.05	This team (2)
	Nigeria	4.0	77	301			0.17	This team (3)
Mussel	Morocco					0.5	0.07	Chafik <i>et al.</i> , 1998
	Ghana	2.0	18	65				Biney, 1991
	Ghana	1.6	5.2	130	1.7		0.13	This team (2)
Legal limits (4)		70	100			1.0	0.5	Irwin <i>et al.</i> , 1997

Notes:

1. In order to allow comparison with literature data, it was necessary to use mean values instead of medians, and another biomass unit, namely fresh weight instead of dry weight.
2. Otchere *et al.*, in press.
3. Joiris and Azokwu, 1999.
4. Usual limits, in most countries.

Table 1. Average heavy metal concentrations in some African bivalves ($\mu\text{g/g}$ fresh weight)

A first analysis of such data concerns orders of magnitude, and reveals that in general, essential metals show similar levels in the different bivalves and the different African regions, as to be expected since these metals are submitted to efficient regulation mechanisms. This is clearly the case for manganese (Mn) and for copper (Cu), with the exception of Ivory Coast. Striking is the different behaviour of zinc (Zn): important differences can be noticed between bivalves, the oysters accumulating much more Zn and Cu than the two other species. This seems a strange phenomenon, for two species with apparently similar ecology and feeding behaviour, i.e. filtration. Moreover, geographical differences can be detected as well, with higher Zn concentration in Ivory Coast again, and iron (Fe) in Nigeria.

The contamination of these bivalves depends on other factors, that must be taken into account before making more detailed conclusions. In order to illustrate this, some of our data are presented in Table 2. Geographical differences could be detected, with higher

Zn concentration in the oysters from Benya during both seasons. Seasonal variations, on the other hand, could be detected as well, apparently due to hydrological differences like the effect of rain on draining heavy metals from land, or differences between open and close lagoons, but also to physiological variations, e.g. in reproduction cycle, growth of the gonads, etc. The first conclusion is thus that data collected in coastal areas during the dry and wet seasons cannot be directly compared and should be treated separately.

Season	Species	Location	n	length	Cu	Zn	Fe	Mn	Cd
Dry season	Cockle	Benya	20	29	8.2	36	790	4.8	0.90
		Ningo	10	53	3.0	36	210	15	0.34
		Sakumo	10	29	8.2	5.8	520	18	1.1
	Oyster	Benya	20	47	17	2350	350	11	0.74
		Ningo	10	32	74	380	450	20	1.1
		Sakumo	10	45	40	630	280	17	0.91
Mussel	Benya	20	40	15	16	900	12	1.4	
	Sakumo	10	39	16	12	1130	15	1.9	
Wet season	Cockle	Benya	20	36	4.6	100	1100	9.6	0.30
		Ningo	10	42	4.3	42	830	7.7	0.19
		Sakumo	10	37	4.1	35	570	19	0.13
	Oyster	Benya	15	32	17	2800	550	13	0.21
		Ningo	10	32	59	560	700	13	0.18
		Sakumo	10	43	33	430	525	20	0.12
	Mussel	Benya	15	40	8.4	87	1050	18	0.42
		Sakumo	10	38	7.1	47	610	5.1	0.11

Cockle: *Anadara (Senilia) senilis*; oyster: *Crassostrea tulipa*; mussel: *Perna perna*.

This team: Otchere *et al.*, in press.

Table 2. Seasonal and geographical variations in heavy metal concentration ($\mu\text{g/g}$ dry weight) of total Cu, Zn, Fe, Mn and Cd, and shell length (mm) in bivalves from Ghana: Benya and Ningo are open lagoons and Sakumo is a closed lagoon; median values; n = number of samples.

The age effect is an important factor is: some organisms show differences in stable residue concentrations with age, one of the most typical situations being a slow increase of concentrations with age, e.g. in fish and marine mammals. In some molluscs, however, concentrations can decrease in response to a "growth dilution" effect. In an initial simple approach, the age effect can be detected by expressing concentrations as a function of length in animals that grow throughout their lives, like molluscs and fish. For other animals, that reach an adult stable size, e.g. mammals and birds, actual age should be determined (on the basis of teeth in marine mammals, etc.). Differences in size (age) distribution of the samples might thus affect the concentration of stable residues, and so lead to erroneous conclusions. In the case reported in Table 2, apparent geographical differences, e.g. between Benya and Ningo, can in fact be fully explained by the size effect, as illustrated in Figure 1 for Cu, as an example. Data must thus be normalized for the main factors influencing the accumulation processes, before

embarking on more detailed discussion. In addition to age, these factors include sex, lipid content, diet etc. Actually, the contamination level of an ecosystem can be described by a curve (or its equation) as shown in Figure 1, instead of median or mean concentration values.

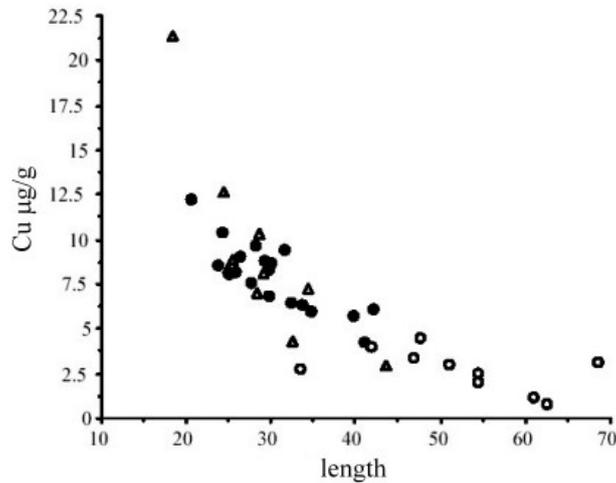


Figure 1. Relationship between total Cu concentration ($\mu\text{g/g dw}$) and length (mm) of cockles from Ghana (dry season); dots: Benya, circles: Ningo, triangles: Sakumo lagoon. See legend to Table 2.

It should be noted that the distribution of pollutant concentration is not normal. Data should, therefore, not be expressed as mean and standard deviation, but as median. Non-parametric statistics only should be applied. Unfortunately, many publications still make use of mean values, and should therefore be treated with great caution.

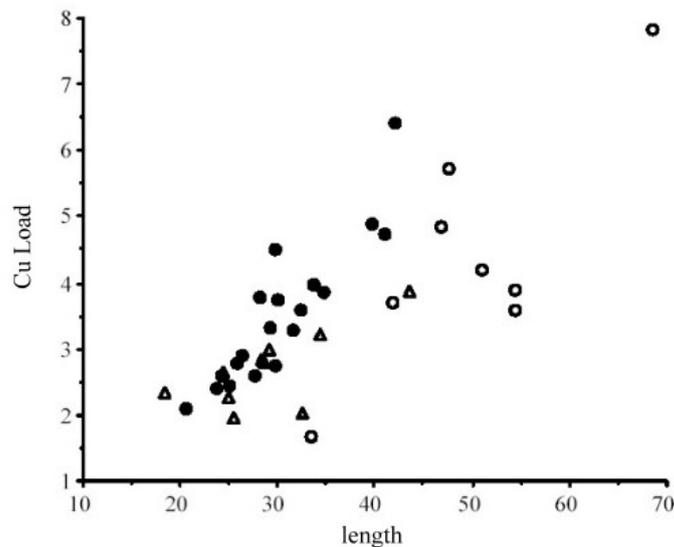


Figure 2. Relationship between total Cu load or burden ($\mu\text{g/ animal}$) and length of cockles from Ghana. See legend to Figure 1.

Such data might suggest that contamination is decreasing with age. In order to test this, it is, however, necessary to express data in other units again, namely in total body load (or burden), by multiplying the concentration expressed as amount per biomass unit, by total body (flesh) weight. It then appears that contamination actually increases with age (See Figure 2): the decrease in concentration just reflects the fact that the growth of the animals is more rapid than the accumulation of metals over time, leading to the so-called "growth dilution effect".

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Biographical Sketch

Claude R. Joiris was born in Etterbeek (Brussels, Belgium) in December 1942. He quickly became an amateur field naturalist, and was an excellent ornithologist at the age of 15. After his studies as an elementary school teacher, he studied zoology at the French speaking free University of Brussels (ULB), and obtained his PhD in 1970 in molecular biology, namely on specific permeases for amino acids in yeast. He then decided, however, to go back to a broader, less specialized topic: he entered the newly founded Flemish University (VUB) as an assistant, in the field of marine ecology. Next to marine bacteria, his official task, he conducted seabird counts during his expeditions at sea, and later, as a full professor, developed studies on the at-sea distribution of seabirds and marine mammals, especially in

polar regions (European Arctic seas, and the Weddell Sea, Antarctica).

As an ornithologist, he soon became very sensitive to the problems raised by the use of organochlorine pesticides, and published a first special issue on the topic in 1968 (Joiris, C. (ed), 1968. *Les oiseaux et les pesticides. Aves*, **5**: 1-64). He then started a study of contamination of Belgian raptors by organochlorine residues, including the mechanisms involved in transfer and accumulation (Joiris C. and K. Delbeke. 1985. Contamination by PCBs and organochlorine pesticides of Belgian birds of prey, their eggs and their food, 1969-1982. In: Nürnberg, ed. *Pollutants and their Ecotoxicological Significance*. John Wiley & sons. pps 403-414).

He has now been involved in ecotoxicological studies for several years, mainly in the North Sea and the Black Sea, and mainly using marine mammals and seabirds found dead (both stranded and bycatch), in an international approach. Whenever possible, he gives priority to collaboration between specialized teams, including veterinarians for pathology, and ecotoxicologists in order to obtain information on as broad as possible a range of stable contaminants. Part of his team concerns organic residues, both organochlorines and organic mercury. As a typical example, let us cite: Holsbeek L, Joiris CR, Debacker V, Ali BI, Roose P, Nellissen JP, Gobert S, Bouquegneau JM and Bossicart M. 1999. Heavy metals, organochlorines and polycyclic aromatic hydrocarbons in sperm whales stranded in the southern North Sea during the 1994/1995 winter. *Marine Pollution Bulletin* **38**: 304-313.

He has promoted about 10 PhD theses in the field of ecotoxicology.